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# Identification of the sources of local higher human polyaromatic hydrocarbon (PAH) exposure in the regions Menen and Genk-Zuid, and policy interpretation

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# EXTENDED ABSTRACT

***Goal.*** The main goal of the current project was to identify the sources of population exposure to polycyclic aromatic hydrocarbons (PAH) in the regions Genk/Diepenbeek and Menen (Flanders), and guiding the translation of the results into policy actions. The study included sampling of PAHs in air on different locations within both study regions. During the sampling periods, qualitative estimation of PAH uptake via food (using questionnaires) and longitudinal analysis of urinary PAK metabolites (1-OH pyrene and OH-phenanthrenes) was executed in a limited study population living near to the sampling locations.

***Materials and Methods.*** Air PAH and $PM_{2.5}$ levoglucosan sampling was done simultaneously on 13 sampling locations – six in each geographical area, and one on a reference location – during a period of 4 weeks between August 31 and October 1, 2015, and between January 19 and March 5, 2016. The sampling areas Genk/Diepenbeek (East of Flanders) and Menen (West of Flanders) were situated at a distance of about 200 km from each other. The background sampling area Veurne-Houtem was situated 50 km West of Menen, at a distance of 10 km from the North Sea. The air samplers were all located near to habitation i.e. in the backyard or on the balcony of the participants' residences. Sampling of PAHs was done using cartridges filled with the sorbent polydimethyl siloxane–Tenax, (which traps suspended particles and gasses) connected to a constant flow air sampling pump GillAIR3 pump at a flow of about 20L/h. Levoglucosan (LEV), a tracer for biomass burning was collected using $PM_{2.5}$ Harvard impactors equipped with Quartz fiber filters, and connected to a pump with a flow of about 10 L/min. In each of the 8 sampling weeks, all N=41 (warm season) and N=42 (cold season) participants collected morning urine for 1-OH pyrene and OH-phenanthrene analysis. The participants filled out a food frequency questionnaire on food items possibly containing PAHs, and consumed in the 24h time-frame before urine collection. Participants were asked not to consume barbecued, grilled and smoked food, and needed to record it if they had been doing so in the day before urine collection.

Twenty-two different PAHs were measured in the air samples, namely the 16 EPA-PAHs, four methyl-PAHs (2-Me, and 1-Me-naphthalene, 9-Me-phenanthrene, 2-Me-anthracene), benzo(e)pyrene, and picene. All the analyses were performed on a Thermal Desorption-GC-MS. Levoglucosan was measured with gas chromatography in combination with mass spectrometric detection in electron impact mode. 1-OH pyrene and OH-phenanthrenes were analyzed using Ultra Performance Liquid Chromatography (UPLC)-tandem MS.

Different techniques were used for source apportionment of PAHs: calculation of diagnostic ratios based on molecular tracers, and principal component analysis (PCA). The association between air PAH levels and urinary concentrations, was assessed using mixed effects multiple linear regression analysis.

## *Results*

***Study population and home environment***. In total 50 individuals participated in the study, 25 males and 25 females, living on 29 different locations. The average age of the participants was 57.7 y, of which 51% were employed at diverse education levels. All individuals were non-smokers, however 48% indicated ever having smoked. Forty individuals (77.5%) reported activities of industrial and/or traffic origin in the neighborhood of their house, causing increased exposure to combustion products. Some **geographical parameters** were calculated for the home environment of the participants. 13.9% of the residences were situated within a high residential area. They were either located in a busy road with more than 10000 vehicles/day or maximal ca. 1.2 km away from it. The measuring units of the traffic locations of Genk/Diepenbeek (G01, G21, G02 and G29) were located in streets with more than 10000 vehicles/day. Also on the locations G03 (industry) and G06 (background) the nearest road was one with high traffic intensity (resp. 97 and 181 m away). This was a bit different in Menen. The nearest roads to the traffic locations, had much less (heavy) traffic (M05, M06). On the industry locations M01 and M11, there was still quite some traffic intensity in the neighborhood, whereas at industry location M02 there was much less traffic (only 104 vehicles per day on the nearest road, and the nearest major road (>10000 vehicles/day) being about 1.2 km away). At the sampled background residential locations of Menen (M03, M04), there was relatively high traffic intensity nearby (respectively 9686 and 5689 vehicles/day on roads respectively 139 and 173m away), and the nearest major road was about within a distance of 234 and 320m respectively. The surface of nature in a perimeter of 5 km was similar for all measuring locations (range of: 3.1 - 5.6E+07 $m^2$).

***Meteorological conditions during the sampling periods***. The **average daily air temperatures** during the sampling periods in Sept'15 (13.6°C) and Jan-Feb'16 (5.2°C) were comparable with monthly normal temperatures for respectively the months September (14.9°C) and January-February (3.3 and 3.7°C) in the central measuring location of Ukkel (Belgium, period 1981-2010). The median **daily precipitation** was in both seasons 0.20 mm i.e. 0.20 L/$m^2$ i.e. considerably lower than the normal values for the sampling months (4.38, 3.98 and 3.87 mm for September, January and February respectively). During both sampling periods, the levels of $PM_{10}$ and $PM_{2.5}$ measured by the Flemish Environmental Agency (VMM) on nearby stations, were relatively low. The median level in the summer-fall and winter sampling periods (over three sampling locations) were respectively for $PM_{10}$ 14.0 and 17.0 µg/$m^3$, and for $PM_{2.5}$ 7.0 and 10.0 µg/$m^3$. At the start of the winter period (end of January 2016) there was however a short smog period. With $PM_{10}$ levels above the EU 24h guideline value of 50µg/$m^3$. In the second halve of February there were also some days with moderately higher PM levels. Those periods occurred during some colder days in January and February 2016.

***Levels of PAHs and levoglucosan measured in the current study***. Overall, the levels of PAHs were clearly higher in Genk and Menen compared to Houtem, only in the summer-fall period and not during the winter campaign: sum of 22 PAHs were respectively 45.0, 50.1 and 3.6 ng/$m^3$ in summer-fall, compared to 42.2, 31.0 and 54.3 ng/$m^3$ in winter. The largest proportion of that PAH sum was represented by low molecular weight PAHs. This proportion - which was in the range of 78.5% to 92.4% of total PAHs - was slightly increasing in the winter time to the range of 92.2% to 93.9%. The magnitude of the PAH concentrations measured in the current study could approximately be compared with levels observed in previous studies. An exact comparison was not possible, as the sampling techniques used, the seasons sampled, and the (number of) PAH compounds assessed, were different. Comparison with studies, in which both gaseous and particle bound PAHs were analyzed, showed that the range of PAHs observed in the current study were comparable to levels measured in the UK and France. Especially a recent study of Tomaz et al. (2016) covering a year-long (year 2013) monitoring on a measuring air quality station in the city of Grenoble (France) could be used for a more detailed comparison. Comparing the yearly averages of the sum of 17 common PAHs between that study and the current study, showed that the average total PAH levels were very similar (respectively 22.4 in Grenoble vs. 24.5 ng/$m^3$ in the current study). 1MeNAP was an exception, to the Grenoble levels. The concentration in the current study was clearly higher. In Menen the 1MeNAP levels were associated with the surface of industry areas within perimeter of 3 km around the measuring units.

The levoglucosan levels observed in the current study, were as expected, correlated with the lower molecular weight PAHs (r=0.19, p=0.018) measured at the same locations. Indeed, during biomass burning, mainly lower molecular weight PAHs are formed. In general, the levoglucosan concentrations were quite high. The median levels over the different measuring locations were for Genk, Menen and Houtem: 33.2, 44.6 and 24.3 ng/m$^3$ in summer-fall, increasing to 118.5, 210.0 and 87.7 ng/m$^3$ in winter. At all individual measuring locations, the winter values increased with a factor 2 to 7 compared to summer-fall. The contribution of wood burning tot the PM$_{2.5}$ mass varied from less than 10% (mostly in summer-fall) to more than 20%. Although this contribution is considerable, similar wood burning PM$_{10}$ percentages were observed during a levoglucosan measuring campaign performed in the year 2010 on several locations in Flanders (Maenhaut et al., 2012). Recently Maenhaut et al. (2016) estimated that the conversion factor used to calculate the contribution of wood burning to PM mass was twice as high for Flanders, which would mean that in our study the PM$_{2.5}$ mass originating from wood burning would be estimated to be between less than 20 to over 40%.

**Identification of specific PAH sources in Menen, Genk/Diepenbeek and Houtem**. We calculated diagnostic ratios, being the ratio of two or more PAHs, to get an indication of the type of PAH sources present in the regions. The ratio IcdP/(IcdP+BghiP) [1]is used to estimate either petrogenic (ratio<0.2), petroleum combustion (mixed sources, ratio between 0.2 and 1.5) or grass/wood/coal combustion (ratio>0.5) sources. In summer-fall, the ratio IcdP/(IcdP+BghiP) showed combustion origin of the PAHs sampled on the industry and traffic locations of Genk/Diepenbeek (ratio>0.5), whereas petroleum combustion samples (ratio between 0.2-0.5) originated from both the industry and background stations. In summer-fall the IcdP/(IcdP+BghiP) ratios in Menen were mostly above 0.5, and for some background samples between 0.2-0.5. In winter, on all locations were the levels were above detection limit, the IcdP/(IcdP+BghiP) ratios were >0.5 (Genk/Diepenbeek), indicating combustion of biomass and coal as a primary PAH source. This was confirmed by the BaA/(BaA+CHR) ratios which were almost all higher than 0.35 (combustion).

Factor analysis was also performed to perform source apportionment of the PAHs, separate for the location Menen and Genk. As for diagnostic ratios this is a tool indicative for possible sources. Indeed, PAH patterns can differ according to sampling/analysis techniques, mixing up of sources, ageing of the air sample due to atmospheric reactivity of some PAHs with other atmospheric species. Based on the factor analysis, it was observed that both in Genk and Menen, important sources of PAHs were vehicle emissions (5, and 6-ring PAHs), and diesel (FLU, PHE, FLA, ANT, PYR). In the summer period, the background measuring unit M04 showed relatively high levels of ACE and ACY, appearing as separate factor in the factor analysis. These compounds theoretically originate from the steel/iron industry, from burning of heavy oils or possibly petrochemistry. In winter, similar compounds were seen, and a biomass burning factor (including among others NAP, PHE, FLA, PYR, levoglucosan) appeared in the factor analysis, as well as a factor loaded with BaP and CHR, potentially originating from natural gas combustion. In Menen, similar factors came out of the factor analysis, being in summer mainly vehicular emissions, and industry and (light oil) combustion processes the most important potential sources. In winter, wood combustion was also strongly present in the factor analysis (factor loading of 52% explained variance for combined factor of wood burning and traffic emissions).

For identification of specific sources, observation of individual day-to-day and seasonal trends were especially interesting. Increases in levoglucosan were consistently observed in the colder periods (19/1, 21/1, 11/2 and 16/2/16) reaching levels in the range of approximately 300 to 900 ng/m$^3$ on nearly all locations (except industry station G03, and the background station G05). The PAHs showed a more locally diverse pattern. For each of the sampling locations the compounds specifically popping up are indicated in the table below.

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[1] For abbreviations of the PAHs mentioned, see Table 2

| Station | Observations |
|---|---|
| **Genk traffic: G01, G02, G21, G29** | - G01 and G02: rather consistently relatively higher levels of low molecular and high molecular weight PAHs, which is typical for diesel and vehicular emissions. Both stations located in major heavy traffic roads: respectively 17.10$^3$ and >20.10$^3$ vehicles/day, of with around respectively 2000 and 1000 heavy traffic vehicles/day<br>- G21 and G29: replaced respectively G01 and G02 in the winter time. G21 was also located in a major road with heavy traffic (>20.10$^3$ vehicles/day of which around 2000 heavy vehicles/day). Whereas the G29 location was a major road with about 11000 veh/day, but without heavy traffic.<br>- G21 showed rather high and G29 somewhat variable low- to medium-molecular weight PAHs (NAP to PYR). On both locations much lower concentrations of the 5-rings IcdP, DahA, PIC and the 6-ring BghiP were observed, compared to the summer stations G01 and G02.<br>**-> Possible sources:** mainly traffic emissions |
| **Genk Industry: G03, G04** | - less clear to indicate PAH sources<br>- **G03** and **G04**, respectively W and N of the industrial area Genk-South.<br>- both stations had relatively high values of medium to high molecular weight PAHs (4- to 6-rings) in the summer fall campaign. During the winter campaign, G04 did not shown this anymore.<br>- **G03**: on 2 summer-fall days (15/9 and 22/9/15), the 2- and 3-ring PAHs from NAP to PHE were relatively high. In winter all low molecular PAHs were now clearly present at all winter sampling days, possibly originating from (wood) combustion sources.<br>- **G04**: In summer the high ACY and ACE levels were very apparent. Also here 15/9 and 23/9/16 were the days with clearly highest levels of here mainly, ACE and 9MePHE. During the winter campaign this was not observed, although ACY was still visible in the profile.<br>**-> Possible sources:** traffic and some industrial sources. ACY and ACE originating from steel/iron industry which use heavy oils, or possibly petrochemistry? |
| **Genk background: G05** | - similar concentrations of medium to high molecular weight PAHs as observed in the industry monitoring stations G03, and even more similar to G04. G05 also showed tendency of increased ACE and ACY in summer-fall, which was somewhat reduced to a slightly increased tendency of ACY in winter.<br>- Measuring units G04 and G05 were located 2 km away from each other respectively at N and N-E of Genk-South industry. Both in streets without heavy traffic, and with respectively around 4000 and 1500 vehicles/day<br>**-> Possible sources:** some influence of sources on G04 industry location |
| **Menen industry: M01, M02, M11** | - **M01** and **M02:** respectively W and N-W of Menen industrial area. M11 station replaced the M01 station from the summer-fall period, and was located only a few houses apart.<br>- Summer-fall: M01 and M02 showed a similar pattern of relatively high levels of 4- and 5-rings, and higher appearance of low molecular PAHs (NAP, 2MeNAP, 1MeNAP, ACE, PHE) in the first sampling days of the summer-fall campaign (1-10/9/15). In the next sampling period (15/9-24/9), those PAHs were low, and only 9MePHE appeared relatively strong in the PAH spectrum.<br>- Winter: M11 mainly showed the 5- and 6 rings IcdP, DahA, PIC and BghiP, and some lower molecular weight PAHs, in the winter period 9-16/2/16, when levoglucosan was increased. On the location M02, also the lower molecular weight concentrations were higher in the winter campaign compared to the summer-fall period, and also here as for M11, especially in the period of 9-16/2/16, when levoglucosan was increased.<br>- PHE, and mainly 9MePHE visible, mainly in summer, but also in the winter sampling period<br>**->Possible sources:** mixture of vehicular emissions, gasoline combustion, wood and light oil combustion |
| **Menen background:** | - **M03** and **M04** located at the E side of the industry terrains of Menen<br>- in summer-fall relatively high levels of the low molecular PAHs (NAP, 2MeNAP, 1MeNAP, |

| | |
|---|---|
| **M03, M04** | ACE, PHE). In winter period M03 and M04 mainly showed lower molecular weight PAHs, with the highest concentrations in the cold period from 9-16/2/16.<br>- In period 15-24/9/15, also here 9MePHE was clearly present, as was observed for the industry sites M01 and M02<br>**->Possible sources:** vehicular emissions, wood burning, industrial source of low molecular weight PAHs (source of PHE and 9-MePHE?). In Menen 9MePHE was associated with the surface of industry areas within a 3000m perimeter. |
| **Menen traffic: M05, M06** | - **M05**: around 9000 vehicles per day, of which about 600 heavy traffic vehicles per day, all 2-, 3- and 4-ring PAHs were clearly measurable. Before and after the street festival in beginning of September, clearly the low molecular weight and all other PAHs were much lower. It is not probable that this low-PAH period was (only) due to that event.<br>- **M06** was located in a street with less traffic than M05 (6200 veh/day), and mainly peak hour traffic. Due to a technical problem, only little PAH measurements were available in both seasons. For the days the PAHs were available in the summer-fall period, most low molecular weight PAHs were low, except 9MePHE., The 4- and 5-ring PAHs (BaA, BbF, BeP, BaP and IcdP were relatively more present, and to the extend as on the industry locations M01 and M02 and the background station M03.<br>- In the winter time, there were lower PAH levels measured on both traffic locations, except for some days at location M05, e.g. 16/2/16, where clearly higher levels of 2-, 3- and 4-rings were observed, together with high levoglucosan concentrations (608.2 $ng/m^3$).<br>**-> Possible sources:** vehicular emissions of diesel on location M05. At M06 some influence of 9MePHE and higher molecular weight PAHs. |
| **Houtem background: H01** | - In summer, all levels of PAHs were low, compared to the locations Menen and Genk.<br>- in second half of September, the levels of 9MePHE were relatively higher, possibly associated with the higher levoglucosan concentration measured on 25/9/15?<br>- In winter period: higher PAH levels were seen mainly when relatively high levels of levoglucosan were measured<br>**-> Possible sources:** mainly wood burning |

NB: PHE and/or 9MePHE appeared clearly in the profile of M01, M02, M03, M04, M05 and M06. PHE was one of the main compounds emitted, aside from NAP and ACY by a company in the Menen industry area

**Urinary PAH metabolites and the link with outdour ambient PAH exposure of people living at and near to the measuring locations.** Repeated urine samples were collected during in total each of the eight sampling weeks. The median concentration of the urinary PAH OH-metabolites was respectively: 35.5 ng/L (Genk) vs. 37.7 ng/L (Menen) for 1-OH pyrene , and 615.9 ng/L (Genk) vs. 717.8 ng/L (Menen) for the sum of OH-phenanthrenes. In the current study population, respectively 14, 44 and 42% of the individuals were of the age group between 33-49y, 50-60y, or >60y. We did not see a trend in the PAH urinary metabolite levels in this age range. therefore we compared the across-ages median levels with a Flemish (FLEHS-1) adult population between 50 and 65y old. When excluding from the FLEHS-1 population, individuals which either smoked, recently ate barbecued/grilled food, or had indoor heating with a stove, the median(IQR) 1-OH pyrene level of the Flemish population was 30 (30-140) ng/L, which was in the range of the concentrations measured in the current campaign. Similar 1-OH pyrene levels were reported in the same age classes (smokers+non-smokers) in: USA 32.5 ng/L (N=507) (Best et al. 2016), and Australia where urine pools were analysed with levels between 50 and 96 ng/L (Thai et al., 2016).

Based on mixed effects single linear regression models, only sex showed a potential influence on creatinine adjusted 1OH-Pyrene levels. In Menen, the average OH-phenanthrene levels were 1.36 times higher as compared to Genk (p=0.01). Individuals from which the parents were born in Belgium, had 1.84 times higher levels than those who were not (p=0.001).

The OH-metabolite levels were not correlated with the outdoor PAH or levoglucosan concentrations. After adjustment for covariates, there was no significant association between ambient PYR or levoglucosan concentrations and 1-OH pyrene in urine. In the same way, also ambient PHE+ANT or levoglucosan concentrations were not correlated with OH-phenanthrenes in urine. Aquilina et al. (2010) reported no correlation between FLU, PHE or PYR measured via personal samplers and 1-OH pyrene analyzed in urine of 100 non-smoking subjects living in the UK. In a study on non-smoking pregnant women, high density housing, passive smoking exposure, and residential heating with coal showed up as significant predictors of PAH exposure (Polanska et al., 2014). Since intake of PAHs via smoking and food consumption were exclusion criteria and thus limited to a minimum level in the current population, air PAH levels, were speculated to be the main cause for concentration variations seen in individuals. Aside from outdoor air levels, an unknown factor of influence in the current study was exposure via indoor sources, influenced by the way of ventilation of the buildings. Furthermore, individuals also accumulate PAH exposure via exposure in traffic, not only at their home address, but also via use of different transportation modes.

**Policy advices.** The current project lead to formulation of some policy advices on the level of the citizen, local communities and the larger regional level. Citizens can change their behavior to obtain lower exposure to PAHs by avoiding sources of PAHs e.g. no smoking, avoidance consumption of burned food, smart ventilation and airing, application of best practice for barbecuing food and use of woodstoves. Measures that can be taken in specific areas to reduce PAH exposure, can be focused on traffic by improving public transport, improving traffic flows, stimulation of biking, avoidance of heavy truck traffic near to residential areas, communication towards citizens about actual ambient air quality, more efficient use of water ways for industrial traffic (which would be an option both for Menen and Genk/Diepenbeek). Overall on larger Flemish level, green energy should be stimulated to lower burning processes needed for driving residential heating, engines, and industrial plants. Stimulation of electric heating using electricity generated via photovoltaic cells or wind mills, would considerably reduce PAH levels in the direct life environment. In the same way stimulation of traffic means powered by green energy, could lower PAH emissions in residential areas.

***Conclusion.*** Based on increased 1-OH pyrene levels in Menen and Genk, observed during the FLEHS-3 biomonitoring, the current PAH and levoglucosan monitoring campaign was started to assess possible local sources of outdoor PAHs in residential areas of those regions. We could not observe a link between the ambient PAH concentrations and urinary OH metabolites repeatedly measured in the participants, in each of the air sampling weeks. Nevertheless, clearly, Menen and Genk/Diepenbeek had higher concentrations for most of the PAHs compared to the background location Houtem, although this difference was much less apparent in the winter period.

Lower molecular weight PAHs were correlated with levoglucosan levels measured at the same locations, indicating that biomass burning was an important source of those PAHs in all regions. In Genk/Diepenbeek the low molecular weight PAH ACY was popping up in measuring stations near to the industrial area. In Menen 9-Me PHE and 1MeNAP were linked with the presence of industrial areas and heavy traffic. Furthermore, the PAH profiles on most of the measuring locations in Genk/Diepenbeek and Menen were indicative for traffic as one of the important sources generating PAHs in the residences ambient air environment. Overall, air contamination with PAHs remains a matter of concern in whole over Europe and beyond. PAHs are emitted via residential heating, traffic, and industry. Initiatives on personal exposure prevention as well as local or regional reduction measures, fit into the policy of transition to green energy.

# UITGEBREIDE NEDERLANDSTALIGE SAMENVATTING

**Doelstelling.** Het belangrijkste doel van het huidige project was het onderzoek naar de bronnen van blootstelling aan polycyclische aromatische componenten (PAK's) in de regio's Genk/Diepenbeek en Menen en aanzetten tot vertaling van de resultaten in beleidsacties. De studie omvatte een staalnamecampagne van PAK's in de lucht op verschillende locaties in beide regio's. Tijdens de samplingsperiode werd bij de deelnemers, naast luchtmetingen ook via voedingsvragenlijsten ingeschat wat de PAK-opname was via het dagelijks dieet. Bij de deelnemers werden een metaboliet van de PAK pyreen en fenantreen gemeten in urine van een beperkte onderzoekspopulatie die woonden op of in de buurt van de staalnamelocaties.

**Materiaal en methoden.** De luchtsampling werd gelijktijdig uitgevoerd op 13 locaties - zes in elk geografisch gebied, en één op een referentie-locatie. De staalname vond plaats gedurende een periode van 4 weken tussen 31 augustus en 1 oktober 2015 en tussen 19 januari en 5 maart 2016. De bemonsteringsgebieden Genk/Diepenbeek (Limburg) en Menen (West-Vlaanderen) bevonden zich op een afstand van ca. 200 km. Het achtergrondgebied Veurne-Houtem was gelegen op 50 km ten westen van Menen, op een afstand van 10 km van de Noordzee. De luchtsamplers werden opgesteld in de achtertuin of op het balkon van de woningen van de deelnemers. Bemonstering van PAK's werd gedaan in zwevende deeltjes en in de gasfase met behulp van patronen (cartridges) gevuld met het sorbens PolyDimethylSiloxaan-Tenax, aangesloten op een bemonsteringspomp met een debiet van 20L/uur. Levoglucosan (LEV), een tracer voor biomassaverbranding werd verzameld d.m.v. $PM_{2.5}$ Harvard impactor voorzien van een kwartsvezelfilter en verbonden met een pomp met een debiet van ca. 10L/min. In elk van de 8 bemonsteringsweken, verzamelden alle (N=41 en N=42 in resp. zomer-herfst en winter seizoen) deelnemers ochtendurine voor 1-OH pyreen en OH-fenantrenen analyse. De deelnemers vulden een voedselfrequentievragenlijst over consumptie, in de laatste 24 uur voor de urinecollectie, van voedingswaren die potentieel PAK's bevatten. De deelnemers werd gevraagd om geen gebarbecuede, gegrilde en gerookte voedingswaren te eten de dag voor de urinecollectie.
Tweeëntwintig verschillende PAK's werden gemeten in de luchtmonsters: 16 EPA-PAK's, vier methyl-PAKs (2-Me en 1-Me-naftaleen, 9-Me-fenantreen, 2-Me-antraceen), benzo[e]pyreen en piceen. Alle analysen werden uitgevoerd via thermische desorptie gaschromatografie (TD GC-MS). Levoglucosan werd gemeten via gaschromatografie in combinatie met massaspectrometrische detectie in elektronimpact modus. 1-OH pyreen en OH-fenantrenen werden geanalyseerd met Ultra Performance Liquid Chromatography (UPLC)-tandem MS. Verschillende technieken werden gebruikt voor brontoewijzing van PAK's: berekening van diagnostische ratio's op basis van moleculaire tracers, en principale componenten analyse (PCA). De associatie tussen lucht PAK-niveaus en urinaire concentraties, werd berekend met behulp van mixed-effects meervoudige lineaire regressie-analyse.

**Resultaten**

*Studiepopulatie en thuisomgeving*. In totaal 50 individuen namen deel aan het onderzoek, 25 mannen en 25 vrouwen, die woonden op 29 verschillende locaties. De gemiddelde leeftijd van de deelnemers was 57.7 jaar en 51% was beroepsactief. Alle deelnemers waren niet-rokers, maar 48%

gaf aan ooit te hebben gerookt. Veertig personen (77,5%) rapporteerde industriële activiteiten en/of verkeer in de buurt van hun huis, die verhoogde blootstelling aan verbrandingsproducten kon geven. 13,9% van de woningen waren gelegen in dicht bevolkte delen van de stad. Ze lagen in een drukke weg met passage van meer dan 10.000 voertuigen per dag of maximaal op ca. 1.2 km afstand daarvan. De meetstations op de verkeerslocaties van Genk/Diepenbeek (G01, G21, G02 en G29) bevonden zich in straten met passage van meer dan 10.000 voertuigen/dag. Ook op de locaties G03 (industrie) en G06 (achtergrond) had de dichtstbijzijnde weg (op resp. 97 en 181 meter) deze hoge verkeersintensiteit. De situatie was anders in Menen. De dichtstbijzijnde wegen in de buurt van de verkeersstations hadden veel minder (zwaar) verkeer (M05, M06). De industrie meetstations M01 en M11, waren wel gelegen in straten met veel verkeerspassage, terwijl op de industrielocatie M02 er veel minder passage was van voertuigen (slechts 104 voertuigen per dag op de dichtstbijzijnde weg, en de dichtstbijzijnde grote weg met >10.000 voertuigen/dag lag op ongeveer 1.2 km afstand). Op de achtergrond woonlocaties van Menen (M03, M04), was de verkeersintensiteit relatief hoog: respectievelijk 9686 en 5689 voertuigen/dag op de wegen op 139 en 173m afstand. De dichtstbijzijnde grote wegen lagen binnen een afstand van respectievelijk 234 en 320m. De oppervlakte van groene ruimte in een straal van 5 km, was quasi gelijk voor alle meetlocaties (3.1-5.6*$10^7$ $m^2$).

*Meteorologische omstandigheden tijdens de bemonstering periodes*. De gemiddelde dagelijkse luchttemperatuur tijdens de bemonsteringsperiodes in Sept'15 (13,6°C) en Jan-Feb'16 (5,2°C) was vergelijkbaar met de maandelijkse normale temperatuur voor respectievelijk de maanden september (14,9°C) en januari-februari (3,3 en 3,7°C) (gemeten in de centrale meetplaats Ukkel (België), periode 1981-2010). De mediane dagelijkse neerslag was in beide seizoenen 0.20 mm (0.20L/$m^2$), wat aanzienlijk lager was dan de normaalwaarden voor deze bemonsteringsmaanden (4,38, 3,98 en 3,87 mm voor september, januari en februari respectievelijk). Tijdens beide bemonsteringsperiodes, lagen de niveaus van $PM_{10}$ en $PM_{2.5}$ relatief laag (gemeten door de Vlaamse Milieumaatschappij (VMM) op nabijgelegen meetstations). De mediaangehalten (over drie bemonsteringslocaties) waren respectievelijk voor $PM_{10}$ 14.0 en 17.0 µg/$m^3$, en 7.0 en 10.0 µg/$m^3$ voor $PM_{2.5}$ (voor respectievelijk de zomer-herfst en winter bemonsteringsperioden). Bij aanvang van de winter bemonsteringsperiode (eind januari 2016) was er een korte smog- periode met $PM_{10}$ niveaus boven het EU-24u richtwaarde van 50 µg/$m^3$. In de tweede helft van februari waren er ook enkele dagen met matig hogere PM niveaus. Die periodes vielen samen met koudere dagen in januari en februari 2016.

*Niveaus van PAK's en levoglucosan gemeten in de huidige studie*. In de zomer-herfst periode, maar niet in de winter, waren de PAK's luchtgehalten duidelijk hoger in Genk en Menen in vergelijking met het achtergrondgebied Houtem: som van de 22 PAK's respectievelijk 45.0, 50.1 en 3.6 ng/$m^3$ in de zomer-herfst t.o.v. 42.2, 31.0 en 54.3 ng/$m^3$ in de winter. Het grootste deel van de PAK-concentratie werd vertegenwoordigd door laag moleculair gewicht PAK's. Het aandeel van laag moleculair gewicht PAK's 78.5% tot 92.4% van de totale PAK's in de zomer, nam toe tot 92.2% tot 93.9% tijdens de wintercampagne. De PAK's gehalten gemeten in de huidige studie waren vergelijkbaar met eerdere studies. Een exacte vergelijking was echter niet mogelijk, omdat in andere studies andere staalnamemethodes werden gebruikt en de seizoenen waarin bemonsterd en het aantal PAK componenten gemeten, vaak verschillend was. De waarden gemeten in de huidige studie was vergelijkbaar met metingen in het Verenigd Koninkrijk en Frankrijk, waar - zoals in de huidige studie - gasvormige en deeltjesgebonden PAK's samen werden bemonsterd. Vooral een recente studie van Tomaz et al. (2016) gaf interessant vergelijkingsmateriaal. Een jaar lang (gedurende 2013) werden PAK's gemeten op en luchtkwaliteitsstation in de stad Grenoble (Frankrijk). De jaargemiddelde niveaus van 17 PAK's gemeten in de huidige (22.4 ng/$m^3$) en in de 'Grenoble' (24.5 ng/$m^3$) studie waren erg vergelijkbaar. De PAK 1-methyl naftaleen (1MeNAP) was echter in de huidige studie duidelijk hoger in vergelijking met de Grenoble studie. In Menen was de

concentratie 1MeNAP gemeten op de meetposten gecorreleerd met de oppervlakte industriegebied in een perimeter van 3 km rond het meetstation.
De levoglucosan niveaus waargenomen in de huidige studie waren, zoals verwacht, gecorreleerd met de laagmoleculaire PAK's gemeten op dezelfde locaties (Spearman rank $r=0.19$, $p=0.018$). Tijdens de verbranding van biomassa worden inderdaad vooral lager molecuulgewicht PAK's gevormd. Algemeen lagen de levoglucosanconcentraties vrij hoog. De mediane niveaus over de verschillende meetlocaties waren voor Genk, Menen en Houtem: 33.2, 44.6 en 24.3 ng/m$^3$ in de zomer-herfst, oplopend tot 118.5, 210.0 en 87.7 ng/m$^3$ in de winter. Op alle meetlocaties, waren de winterwaarden een factor 2 tot 7 hoger in vergelijking met de zomerwaarden. De bijdrage van de verbranding van hout tot de $PM_{2.5}$ massa varieerde van minder dan 10% (vooral in de zomer-herfst) tot meer dan 20%. Dit is een aanzienlijke bijdrage, maar soortgelijke contributies werden geobserveerd in het jaar 2010 op verschillende locaties in Vlaanderen (Maenhaut et al., 2012). Onlangs rapporteerde Maenhaut et al. (2016) dat de factor gebruikt om de bijdrage van biomassa aan de PM massa te berekenen, voor Vlaanderen twee keer zo hoog is dan eerder aangenomen. Dit zou betekenen dat in onze studie verbranding van hout voor 20% tot meer dan 40% bijdraagt aan de $PM_{2.5}$ massa.

*Identificatie van specifieke PAK's bronnen in Menen, Genk/Diepenbeek en Houtem*. In de huidige studie werden diagnostische verhoudingen van PAK's berekend. Dit is de ratio van twee of meer PAK's, die een indicatie geven van het type PAK bronnen. De verhouding IcdP/(IcdP+BghiP)[2] wordt gebruikt om in te schatten of de bron petrogeen (petroleumgerelateerd) is (ratio <0.2), of het gaat over een gemengde bron van aardolieverbrandingsproducten (ratio tussen 0.2 en 1.5) of de bron van PAK's eerder gras/hout/steenkoolverbranding is (verhouding>0.5). In de zomer-herfst periode was de ratio van IcdP/(IcdP+BghiP) gemeten op de industrie- en verkeerslocaties van Genk/Diepenbeek groter dan 0.5. Op de industrie- en achtergrondstations van Genk/Diepenbeek waren de ratio's indicatief voor aardolieverbrandingsproducten (ratio tussen 0.2-0.5). In de zomer-herfst periode lagen de IcdP/(IcdP+BghiP) verhoudingen in Menen meestal boven 0.5 en voor sommige achtergrondmonsters tussen 0.2-0.5. In de winter lag de ratio in Genk/ Diepenbeek steeds boven 0.5, wat indicatief was voor verbranding van biomassa en kolen als primaire bron (NB: in Menen ratio niet berekenbaar in de winter door waarden onder detectieniveau). Dit werd bevestigd door de BaA/(BaA+CHR) ratio's die bijna allemaal hoger waren dan 0.35, wat eveneens een diagnostische ratio score is die wijst op verbranding als oorzaak voor de gemeten PAK's.

Factoranalyse werd ook uitgevoerd om een inschatting te krijgen van de bronnen van PAK's in Menen en Genk. Zoals bij diagnostische ratio's is dit een berekeningsmethode die slechts indicatief is voor mogelijke PAK's bronnen. Inderdaad, kunnen de verhoudingen en de aard van PAK's die worden gemeten worden bepaald door de bemonstering/analysetechnieken, het opmengen van verschillende bronnen (die elk bepaalde PAK's genereren) en veroudering van het luchtmonster door atmosferische reactiviteit van een aantal PAK's met andere atmosferische componenten. De factoranalyse leverde voor zowel Genk als Menen, factoren op waarin voertuigemissies en diesel PAK's voorkwamen (FLU, PHE, FLA, ANT, PYR en 5, en 6-ring PAK's). Tijdens de zomerperiode, waren er vooral op de achtergrond meetpost M04 relatief hoge gehalten meetbaar van acetyleen (ACE) en acyteleen (ACY), wat resulteerde in een afzonderlijke factor in de factoranalyse. Deze verbindingen kunnen theoretisch geëmitteerd worden door de staal/ijzer-industrie, via verbranding van zware oliën of eventueel door petrochemische nijverheid. In de winter werden gelijkaardige PAK's, samen met biomassaverbrandings-PAK's gemeten (o.a. NAP, PHE, FLA, PYR, levoglucosan). De factor die benzo[a]pyreen (BaP) en chryseen (CHR) bevatte, was mogelijk afkomstig van de verbranding van aardgas. In Menen, kwamen soortgelijke factoren uit de factoranalyse: in de

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[2] Zie tabel 2, voor afkorting van vermelde PAK's

zomer voornamelijk voertuigemissies, en industrie en/of (lichte olie) verbrandingsprocessen; in de winter, houtverbranding en verkeersemissies.

Voor de identificatie van specifieke bronnen, werden de dag-tot-dag en seizoensgebonden trends nagegaan op de verschillende meetstations van de studie. Sterke stijgingen van levoglucosan werden consistent waargenomen in de koudere perioden (19/1, 21/1, 11/2 en 16/2/16). Ze bereikten dan gehalten gaande van ongeveer 300 tot 900 ng/m$^3$ op bijna alle locaties (behalve industriestation G03, en de achtergrondstation G05). De PAK's componenten vertoonden een meer lokaal divers profiel. De verbindingen die specifiek voorkwamen op elk van de meetstations zijn opgenomen in onderstaande tabel.

| *Station* | *Observations* |
|---|---|
| **Genk verkeer: G01, G02, G21, G29** | - G01 en G02: tamelijk consistent relatief hogere niveaus van laagmoleculaire en hoogmoleculaire PAK's, typisch voor diesel en voertuigen emissies. Beide stations gelegen aan drukke verkeerswegen: respectievelijk 17000 en >20000 voertuigen/dag en respectievelijk 2000 en 1000 zware voertuigen verkeer voertuigen/dag<br>- G21 en G29: vervangingsstations van respectievelijk G01 en G02 in de winter. G21 was ook gelegen in een belangrijke weg met veel verkeer (> 20103 voertuigen/dag, waarvan ongeveer 2000 zware voertuigen/dag). G29 was een locatie met een belangrijke weg van 11000 voertuigen/dag, maar zonder zwaar verkeer.<br>- G21 had vrij hoge gehalten en G29 enigszins variabele lage tot gemiddeld-moleculair gewicht PAK's (NAP tot PYR). Op beide locaties werden veel lagere concentraties van de 5-ringPAK's IcdP, DahA, PIC en de 6-ring BghiP waargenomen, vergeleken met de zomer stations G01 en G02.<br>-> **Mogelijke bronnen:** vooral verkeersemissies |
| **Genk Industrie: G03, G04** | - G03, G04 - minder duidelijke PAK's bronnen<br>- G03 en G04, respectievelijk gelegen ten W en N van het industriegebied Genk-Zuid.<br>- Beide stations hadden betrekkelijk hoge waarden van gemiddeld tot hoog molecuulgewicht PAK's (4- tot 6-ring) in de zomer-herfst campagne. Tijdens de wintercampagne was dit niet meer zichtbaar op meetstation G04.<br>- G03: op 2 zomer-herfstdagen (15/9 en 22/9/15) waren de 2- en 3-ring PAK's van NAP tot PHE relatief hoog. Tijdens de wintercampagne waren alle laag moleculaire PAK's duidelijk aanwezig, mogelijk afkomstig van (hout)verbrandingsbronnen.<br>- G04: In de zomer waren de hoge ACY en ACE niveaus heel duidelijk. Ook hier waren 15/9 en 23/9/16 dagen met duidelijk de hoogste niveaus van vooral, ACE en 9MePHE. Tijdens de wintercampagne werd dit niet waargenomen, hoewel ACY nog steeds zichtbaar in het PAK profiel aanwezig was.<br>**-> Mogelijke bronnen**: verkeer en industriële bronnen. ACY en ACE afkomstig van staal/ijzerindustrie die zware oliën gebruiken of eventueel petrochemie? |
| **Genk achtergrond: G05** | - G04 en G05 lagen op een afstand van 2 km van elkaar, respectievelijk N en N-O van Genk-Zuid-industrie. Beide meetstations lagen in straten zonder zwaar verkeer en met respectievelijk ongeveer 4000 en 1500 voertuigen/dag<br>- Vergelijkbare concentraties van gemiddeld- tot hoog-moleculair gewicht PAK's zoals waargenomen op de industriemeetstations G03 en vooral G04. G05 toonde ook een trend van hogere ACE en ACY concentraties in de zomer-herfst campagne, tot enigszins minder zichtbare aanwezigheid van ACY in de winter.<br>**-> Mogelijke bronnen:** enige invloed van bronnen ook gemeten op de G04 industrielocatie |
| **Menen industrie: M01, M02, M11** | - M01 en M02: respectievelijk gelegen ten W en N-W van het industriegebied in Menen. M11 was het vervangingsstation van M01 in de winterperiode (in dezelfde straat).<br>- Zomer-herfst: M01 en M02 vertoonde een vergelijkbaar patroon van relatief hoge niveaus van 4- en 5-ringen, en hogere concentraties van laagmoleculaire PAK's (NAP, 2MeNAP, 1MeNAP, ACE, PHE) tijdens de eerste bemonsteringsdagen (1-10/9/15). In de volgende bemonsteringsperiode (15/9-24/ 9) waren die PAK's laag en enkel 9MePHE was nog relatief sterk aanwezig in de PAH spectrum. |

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|---|---|
| | - Winter: M11 vertoonde vooral de 5- en 6 ringen IcdP, DahA, PIC en BghiP en sommige lager moleculair gewicht PAK's in de winterperiode 9-16/2/16, wanneer ook de levoglucosanwaarden hoog waren. Op de locatie M02, waren ook de concentraties van lager moleculair gewicht PAK's hoger in de winter in vergelijking met de zomer-herfstperiode. Idem voor M02, vooral in de periode van 9-16/2/16, wanneer levoglucosan verhoogd was.<br>- PHE en vooral 9MePHE zichtbaar in het PAK's profiel vooral in de zomer-herfst, maar ook in de winter bemonsteringsperiode<br>**-> Mogelijke bronnen**: mix van voertuigenemissies, benzineverbranding, hout en lichte olie verbranding |
| **Menen achtergrond: M03, M04** | - M03 en M04 gelegen aan de Oostkant van de industrieterreinen van Menen<br>- In de zomer-herfst relatief hoge niveaus van de lage moleculair gewicht PAK's (NAP, 2MeNAP, 1MeNAP, ACE, PHE). In winterperiode hadden M03 en M04 voornamelijk lagere moleculair gewicht PAK's in het profiel, met de hoogste concentraties in de koude periode 9-16/2/16.<br>- In de periode 15-24/9/15 ook hier 9MePHE duidelijk aanwezig, zoals werd waargenomen voor de industrie meetstations M01 en M02<br>**-> Mogelijke bronnen:** uitstoot voertuigen, houtverbranding, industriële bron van laag moleculair gewicht PAK's (bron van PHE en 9-MePHE?). In Menen was de concentratie van 9MePHE geassocieerd met de oppervlakte van de industriegebieden binnen een perimeter van 3000m. |
| **Menen verkeer: M05, M06** | - M05: passage van ongeveer 9000 voertuigen per dag, waarvan ongeveer 600 zwaar verkeer voertuigen per dag<br>- Alle 2-, 3- en 4-ring PAK's waren duidelijk meetbaar. Voor en na het straatfestival begin september 2015 waren duidelijk lagere gehalten van laag moleculair gewicht en alle andere PAK's meetbaar. Het is niet waarschijnlijk dat dit (alleen) het gevolg was van die gebeurtenis.<br>- M06 was gelegen in een straat met minder (doorgaand) verkeer dan M05 (6200 voertuigen/dag) en met vooral piekuurverkeer. Door een technisch probleem waren er voor M06 weinig PAK metingen bruikbaar in beide seizoenen. Voor de dagen waarop PAK's metingen beschikbaar waren in de zomer-herfst periode, lagen de meeste lage moleculair gewicht PAK's laag, met uitzondering van 9MePHE. De 4- en 5-ring PAK's (BaA, BbF, BeP, BaP en IcdP) waren relatief meer aanwezig, in de mate zoals geobserveerd op de industriemeetstations M01 en M02 en het achtergrondstation M03.<br>- In de winter werden lagere PAK's gehalten gemeten op beide verkeerslocaties met uitzondering van enkele dagen op locatie M05, bv. 16/2/16, waarbij duidelijk hogere gehalten aan 2-, 3- en 4-ringen werden waargenomen en ook hogere levoglucosanconcentraties (608.2 ng/m$^3$).<br>**-> Mogelijke bronnen:** voertuigen emissies van diesel op locatie M05. Bij M06 enige invloed van 9MePHE en hoger moleculair gewicht PAK's. |
| **Houtem achtergrond: H01** | - In de zomer-herfst campagnes waren alle PAK's laag in vergelijking met de locaties Menen en Genk.<br>- In de tweede helft van september, lag het niveau van 9MePHE relatief hoger, mogelijk verband houdend met de hogere levoglucosanconcentratie gemeten op 25/9/15?<br>- In de winterperiode: hogere PAK-niveaus werden vooral gemeten als ook een relatief hoge niveaus van levoglucosan werden gemeten<br>**-> Mogelijke bronnen: vooral hout branden** |

*NB: PHE en /of 9MePHE was duidelijk in het PAK profiel van de meetstations M01, M02, M03, M04, M05 en M06. PHE, naast NAP en ACY, was een van de belangrijkste verbindingen uitgestoten door een bedrijf op het industrieterrein van Menen.*

*Urine-PAK metabolieten en de link met omgevings-PAK blootstelling van mensen die op en in de buurt van de meetlocaties woonden*. Herhaalde urinemonsters werden verzameld gedurende in totaal elk van de acht weken bemonstering. De mediane concentratie van de urine PAK OH-metabolieten was voor 1-OH pyreen respectievelijk 35.5 ng/L (Genk) vs. 37.7 ng/L (Menen) en voor

OH-fenantrenen 615.9 ng/L (Genk) vs. 717.8 ng/L (Menen). In de huidige populatie kwamen respectievelijk 14, 44 en 42% van de individuen uit de leeftijdsgroep tussen 33-49, 50-60 of >60 jaar. We zagen geen trend in de PAK urinaire metaboliet niveaus over deze leeftijdscategorieën. Daarom vergeleken we de waarde van onze studiegroep me een Vlaamse volwassen bevolking tussen 50 en 65 jaar oud (gerekruteerd in 2005-6 tijdens FLEHS-1 humane biomonitoring). We toetsten onze waarden af aan de individuen uit de FLEHS-1 studie die niet rookten, geen geroosterde/gegrilde gerechten hadden geconsumeerd, geen binnenhuisverwarming hadden met stoof of open vuur.  De mediaan (IQR) van 1-OH pyreen urinaire gehalten van die Vlaamse bevolkingsgroep was gelijk aan 30 (30-140) ng/L.  Onze waarden waren m.a.w. vergelijkbaar met die Vlaamse vergelijkingsgroep. Gelijkaardige 1-OH pyreen niveaus werden gerapporteerd in dezelfde leeftijdscategorieën (rokers + niet-rokers) in: USA 32.5 ng/L (N= 507) (Best et al 2016.) en in Australië, met urinaire gehalten tussen 50 en 96 ng/L (Thai et al., 2016).

In Menen waren de gemiddelde OH-fenantreen niveaus 1,36 maal hoger in vergelijking met Genk (p=0.01). Individuen waarvan de ouders geboren waren in België, hadden gemiddeld 1,84 keer hoger niveaus dan degenen die niet van Belgische origine waren (p=0.001). De OH-metabolieten niveaus waren niet gecorreleerd met de buitenlucht PAK of levoglucosan concentraties. Na correctie voor covariaten, was er geen significante associatie tussen omgevingsgehalten aan pyreen of levoglucosan en 1-OH pyreen in urine. Op dezelfde wijze was er geen associatie tussen buitenlucht fenantreen+antraceen gehalten of levoglucosan en OH-fenantrenen gehalten in urine. In dezelfde lijn, rapporteerde Aquilina et al. (2010) geen correlatie tussen FLU, PHE of PYR gemeten via personal samplers en 1-OH pyreen in de urine van 100 niet-rokers uit het Verenigd Koninkrijk. In een studie bij niet-rokende zwangere vrouwen, blekende de factoren ‘hoge dichtheid huisvesting’, ‘passief roken blootstelling’, en ‘verwarming met kolen’ de belangrijke invloedsfactoren voor blootstelling aan PAK’s (Polanska et al., 2014). Aangezien de inname van PAK's via roken en voedselconsumptie uitsluitingscriteria waren in de huidige studie, werd vooraf gespeculeerd dat de belangrijkste oorzaak voor de concentratievariaties bij de deelnemers afkomstig zouden zijn van PAK concentratievariaties in de buitenlucht. Afgezien van de buitenluchtniveaus, was er uiteraard blootstelling in het binnenshuis, blootstelling bij deelname aan het verkeer (evt. in voertuigen).

*Beleidsadviezen*. Het huidige project leidde tot de formulering van een aantal beleidsadviezen op het niveau van de burger, de lokale overheden en/of het Vlaams regionaal niveau. Burgers kunnen hun blootstelling aan PAK's verminderen door het vermijden van bronnen van PAK’s zoals sigarettenrookblootstelling, consumptie van verbrand voedsel, door slimme ventilatie en verluchting en door toepassing van de beste praktijken bij barbecueën en bij gebruik van houtkachels. Maatregelen die lokaal kunnen worden genomen zijn: het beperken van verkeer door het optimaliseren van het openbaar vervoer, stimuleren van het fietsgebruik en vermijden van zwaar vrachtverkeer in de buurt van woonwijken. Verder kan communicatie over de actuele luchtkwaliteit  burgers stimuleren om ook mee te helpen aan de reductie van PAK’s.  Op Vlaams niveau, kan het volop inzetten op groene energie een belangrijke bijdrage leveren om de uitstoot van PAK’s door verkeer, motoren, en industriële installaties te verminderen. Gebouwverwarming is een belangrijke bron van PAK’s. Aanmoediging van het gebruik van verwarmingsinstallaties aangedreven door elektriciteit opgewekt via zonnecellen of windmolens, zou een aanzienlijke vermindering van de PAK-niveaus in de directe leefomgeving kunnen opleveren.

**Conclusie.** Op basis van de verhoogde 1-OH pyreen gehalten die in een eerdere studie werden waargenomen in Menen en Genk (FLEHS-3 humane biomonitoring), werd de huidige PAK en levoglucosan meetcampagne uitgevoerd, om mogelijke lokale bronnen van buitenlucht PAK's te beoordelen in woonwijken van deze regio's. We konden geen verband waarnemen tussen de luchtconcentraties van PAK’s en urinaire OH-PAK metabolieten gemeten in de deelnemers. Niettemin, hadden Menen en Genk/Diepenbeek hogere concentraties van de meeste gemeten PAK’s in vergelijking met de achtergrondlocatie Houtem, hoewel dit verschil veel minder duidelijk was in de winterperiode. Lager molecuulgewicht PAK’s werden gecorreleerd met levoglucosanniveaus gemeten op dezelfde plaatsen, hetgeen aangeeft dat verbranding van biomassa een belangrijke bron van PAK’s was in alle gebieden. In Genk/Diepenbeek was de laag moleculair gewicht PAK ACY duidelijk meetbaar in de buurt van het industriegebied. In Menen bleek 9-MePHE en 1MeNAP gerelateerd met aanwezigheid van industrie en zwaar verkeer. Bovendien waren de buitenlucht PAK-profielen op de meeste meetlocaties in Genk/Diepenbeek en Menen indicatief voor verkeer als een van de belangrijkste PAK genererende bronnen in de woonomgevingen van de deelnemers. Het kan gesteld worden dat luchtverontreiniging met PAK's een punt van zorg blijft in geheel Europa en daarbuiten. PAK's worden uitgestoten via residentiële verwarming, verkeer en bij industriële activiteiten. Initiatieven om persoonlijke blootstelling te verminderen en lokale of regionale maatregelen gericht op het verminderen van PAK’s blootstelling passen in het beleid van transitie naar groene energie.

# CHAPTER 1 INTRODUCTION

In the Flanders Environment and Health (FLEHS II) human biomonitoring campaign of the Centre for Environment and Health (2007-2011) the urinary levels of 1-OH pyrene (a Polycyclic Aromatic Hydrocarbon (PAH) metabolite) measured in youngsters were elevated among the inhabitants of the industrial hotspot areas Genk South (+33%) and Menen (+49%) compared to a reference population in Flanders. Pyrene is a high molecular PAH compound, which is often used as an indicator of the PAH mixture, consisting in itself of several hundred compounds/congeners (about 500 in the air mixture, WHO, 2000[3]).

In both areas, concrete policy actions for both regions were set up by the Flemish Government via a step by step participatory process (expert consultation, stakeholder consultation, focus groups) (Morrens et al., 2013). The current project was the result of a policy action, namely: 'Determining the relative importance of the different uptake routes of PAHs for further policy translation of the established local higher human PAH load'.

PAHs are a class of components that are characterized by fused aromatic rings which are formed by incomplete combustion of organic material. PAHs are formed in complex mixtures of combustion products such as diesel/gasoline exhaust, soot, tobacco smoke, biomass burning and also burnt/smoked/roasted/grilled foods. PAHs can form very complex structures and may exist in a number of isoforms. The components are semi-volatile to slightly volatile. This is amongst others dependent on their molecular weight (MW). The heavier PAHs (MW> 228) attach easily to particles. The number of aromatic rings (from 2 to 10 rings) not only determines the volatility, but also the hydrophobic character of PAHs. If the molecule has more aromatic rings, the water solubility is lower. PAHs can have a biogenic or anthropogenic origin. Biogenic sources of PAHs are scarce. Some PAHs can be created in very small quantities by certain organisms. Furthermore, certain PAHs can be formed in the degradation of terpenes, pigments and steroids. Chrysene (4-ring) may have a biogenic origin. Namely, it is formed in the microbial degradation of pentacyclic triterpenes, which constitute a major component of the wax layer in (land) plants.

The main share of PAHs is of anthropogenic origin. In order to determine what the sources are, molecular profiles of PAHs and other congeners tracer components are measured. Fingerprinting of these components may give an indication of the PAHs origin.

**Petrogenic PAHs** are associated with petroleum (products). They are derived from petroleum products, and are formed by low long-sustained temperatures of 100-300°C. Both crude oil and refined products containing 2- to 4-ring PAHs (naphthalenes, fluorenes, phenanthrenes, chrysenes and dibenzothiophenes). With the exception of the chrysenes, 4- to 6-ring PAHs are almost non-existent in oil products. Characteristic of petroleum products is that the parent PAHs occur in smaller concentrations than the alkylated homologues.

**Pyrogenic PAHs** are aromatic hydrocarbons associated with burning oil, wood, coal (incl. creosote and coal tar). Pyrogenic PAHs are formed during high-speed, high-temperature processes (> 700°C) of engines (vehicles), marine engines and combustion of fuels in coal plants or oil plants. Pyrogenic

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[3] http://www.euro.who.int/__data/assets/pdf_file/0015/123063/AQG2ndEd_5_9PAH.pdf

PAHs also occur at intermediate temperatures (400-600°C) achieved in the processing of coal to coal tar (products) (e.g. creosote, coal tar used in aluminum smelters) and during asphalt paving (Zhang et al., 2015). The pyrogenic PAHs are less alkylated and contain more aromatic rings compared to the petrogenic PAHs. Indeed, pyrogenic PAHs are dominated by 4- to 6-ring PAHs (fluoranthenes, pyrenes, benzo(a)pyrene, benzo(ghi)perylene and indeno(123cd)pyrene). Also 3-ring PAHs, viz. anthracenes and phenanthrenes may be formed during combustion processes. During low-temperature processes (such as wood burning) generally these low molecular weight PAHs are formed. At higher temperatures (such as fuel combustion in engines) organic compounds are cracked into smaller volatile hydrocarbon fragments (pyrolysis), which then form stable PAHs through radical reactions during pyrosynthesis (Tobiszewski and Namiesnik 2012).

In the general population the major routes of exposure to PAHs are intake through diet and through inhalation of polluted air.

1-OH pyrene has been analyzed in all preceding Flemish Environmental Health Surveys (FLEHS). The 4-benzene-ring pyrene metabolite 1-OH pyrene is widely used in urine as proxy for other PAHs present. It is being debated as to which extend it reflects PAH concentrations in air. 1-OH pyrene is thought to mimic the uptake of particle-bound PAHs, through both the lungs and the skin (Rosella et al., 2009). Shin et al. (2013) evaluated environmental PAH modeling with biomarker data and concluded that 1-OH pyrene mainly originated of (indoor) inhalation, and to lesser extend of food intake. Recently Miao et al. (2015) showed that commuters travelling by foot, bike, car or truck had significantly higher urinary 1-OH pyrene concentrations compared to non-commuters. However, other studies showed that food appears to be the dominant source of exposure to pyrene (Li et al., 2010; Cirillo et al., 2006). Recently, Rose et al. (2015) reported little evidence of PAH formation during grilling, frying and toasting experiments, whereas barbecuing over charcoals gave increased PAHs in food. Li et al. (2010) advised for studies on non-dietary sources, consumption of smoked, grilled, and barbecued food should be avoided or used as an exclusion criterion to prevent data biasing. Based on the literature some articles report air pollution as an important contributing source of PAH exposure, while others show that food is an important culprit.

1-OH pyrene has a half-life of 6 to 35 hours. The half-life of PAH metabolites depends on the compound and certainly also on the exposure route, being: 6.0-29.0 h after inhalation exposure; 4.4-12h or 3.1-5.5h after ingestion exposure, and 11.5-15h for dermal absorption (numbers cited in and reported by Li et al., 2012). Morning void urine minimizes intake of PAH from food, and may rather reflect PAH concentrations from indoor and outdoor air. This means that indoor air may also play an important role and might be a confounder of influence in some cases.

The current study focused on assessing PAHs in air of the life environment of the participants, and PAH intake via food estimated qualitatively based on questionnaires. More specifically, this study aimed: (i) to identify regional (specific) PAH sources based on outdoor air PAHs and wood burning tracer patterns. (ii) to assess the influence of the regional PAHs outdoor/food exposure on the urinary PAH levels.

# CHAPTER 2 MATERIALS AND METHODS

## 2.1. AIR SAMPLING SITES AND PERIOD OF SAMPLING

The Sampling areas Genk/Diepenbeek (East of Flanders) and Menen (West of Flanders) were situated at a distance of about 200 km from each other. The background sampling area Veurne-Houtem (further called Houtem) was situated 50 km West of Menen, at a distance of 10 km from the North Sea. PAH and levoglucosan (LEV) air sampling was done at three major types of sampling locations, potentially representing different PAH sources[4]: (i) **Urban hot spot**: addresses within 50m of a road with more than 10 000 vehicles per day (criterion used in EU-FP7 project ESCAPE; traffic data counts from 2007), with exclusion of addresses within 500m of an industrial area. (ii) **Residential near to industry**: addresses within a 200m or 200-500m buffer from the boarder of industrial areas (based on the 'gewestplannen'). (iii) **Residential background:** residential areas with exclusion of addresses within 500m buffer around industrial areas, and exclusion of addresses within 300m buffer around roads with more than 10 000 vehicles per day, and 50m from roads with more than 3000 vehicles per day (traffic counts of year 2007). In both, Menen and Genk/Diepenbeek area, two addresses were selected in each of the three types of sampling locations, resulting in, in total 12 sampling addresses. In Veurne-Houtem one background sampling location was selected.

The air samplers were all located near to habitation i.e. in the backyard or on the balcony of the participants' residences. The inlet of the samplers was placed slightly above persons' height of ca. 2-2.5m. The sampling was done both in the warm and cold season of 2015-2016 i.e. between August 31 and October 1, 2015, and between January 19 and March 5, 2016. At three of the sampling locations (two in Genk/Diepenbeek and one in Menen) there was no repeated sampling in the winter campaign, as the participants of those locations left the study or decided only to collect urine. For those sampling locations, alternative sampling locations were found within maximal 1 km perimeter.

## 2.2. STUDY POPULATION AND INFORMATION ON HOME ADDRESSES

Aside from the participants living on the sampling address, at each of those 12 locations (in Menen and Genk/Diepenbeek), one to three more participants were recruited within a 1 km perimeter. The inclusion criteria were: being in the age range of 50 to 70 years old, non-smoker, living in a non-smokers residence, no occupational contact with PAHs, living at least one year on the address and not moving within the next year, no indoor heating with coal, wood, or open fire. The participants were asked for to avoid grilled, roasted, and barbecued food. In the summer campaign, in total N= 41 people participated. In the winter campaign eight individuals , which left the study, were replaced by nine individuals living in the neighborhood, resulting in N=42 winter campaign participants.

The study was agreed upon by the ethical committee of the University Hospital of Antwerp (Belgian registration nr: B300201525280). All participants were informed about the study and signed an informed consent. At the start of the study, the participants filled out a general questionnaire on

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[4] partly based on the EU guidelines: "Ambient air pollution by polycyclic aromatic hydrocarbons (PAH): position paper, EU, 2001"

socio-economic status (education level, composition of family), health, age, weight, length, possible indoor PAH sources, and housing conditions: type of heating used normally, age of the building, number of rooms, square meters of living area, height of the residence, garage connected to the residence, type of residence.

For each of the addresses of the participants, geographical information was collected on traffic streams, address density and land use at or in a perimeter around the home location. Traffic streams originated from the Flemish multimodal traffic model (base year 2007). Detailed road maps from OpenStreetMap were used to calculate: total road length in a 50m buffer around the residence (‘ROADLENGTH_50’,[m]); traffic intensity on nearest road (‘Q_NEAR’, [vehicles/day]); distance to the nearest road (Q_NEAR) where traffic counts were done (‘DIST_NEAR’, [m]), heavy traffic intensity on the nearest road (‘Q_NEAR_HEAVY’, [vehicles/day]); traffic load being the multiplication of (all or only heavy) traffic intensity with road length in a perimeter of 1000m (respectively ‘TRAFLOAD_1000’ and ‘TRAFLOAD_1000_HEAVY’, [vehicles*m/day]); 1/(distance to the nearest major road) (‘D_NEAR_MAJOR1’, [1/m]); number of bus stops in perimeter of 1000m (‘BUSSTOPS_1000’).

Address densities were point data with static information on addresses for the year 2005 (information from CRAB Light (AGIV)). Other land use variables were obtained from CORINE land cover data for the year 2000 (EEA, 2004). The following variables were calculated: high density residential areas (CORINE Class 111) in a perimeter of 100m (‘HDRES_100’, [yes/no]); Urban green of CORINE class 141, 142 in a perimeter of 3000m (‘UrbGr_3000’, [m2]), Natural land of CORINE class 141, 142, 211, 231, 242, 311, 312, 313, 322, 421, 511, 522 in a perimeter of 5000m (‘NATURE_5000’, [m2])

### 2.3. SAMPLING OF AIR, URINE AND FOOD QUESTIONNAIRES FILLED OUT BY THE PARTICIPANTS

Sampling was done simultaneously on the 13 sampling locations – six in each geographical area, and one on a reference location – during a period of 4 weeks. There were some cases in which the samplers were not operating properly, or participants did not start/stop samplers. In those cases the sampling was done on an alternative nearby day. The weekly sampling scheme was as follows:

At the 13 sampling locations, outdoor air sampling on Tuesday till Thursday and Thursday till Saturday (48h active sampling). Participants living at the sampling locations, filled out a sampling form in which sampling start-stop date and hour were noted down. Sampling of PAHs was done using cartridges filled with the sorbents polydimethyl siloxane(PDMS)-Tenax, (which traps suspended particles and gasses) connected to a constant flow air sampling pump GillAIR3 pump at a flow of about 20L/h (GSA Messgerätebau GmbH). Levoglucosan (LEV), a tracer for biomass burning was collected using $PM_{2.5}$ Harvard impactors equipped with Quartz fiber filters (Whatman Grade QMA 37mm filters). The pumps connected to the Harvard impactors were of the type Tecora, KNF Neuberger N035AN.18, or Harvard, and were set at a flow of about 10 L/min. The flow of all pumps was checked before and after each 4-weeks sampling period. The average pump flow of the measurement before and after sampling, and the exact sampling duration were used for calculating the volume of air passing through the cartridge or filters.

*Figure 1 Equipment installed on each of the measuring locations: PDMS-Tenax sampler (PAH sampling) and Harvard impactor ($PM_{2.5}$) respectively mounted in and on the gray housing, which was attached to a metal stand. Both samplers were connected via rota-sil tubing to the respective pumps placed on the floor (left side of the stand)*

On Thursday of each of the 8 sampling weeks, all N=41 (warm season) and N=42 (cold season) participants collected morning urine for 1-OH pyrene analysis. The participants filled out a short questionnaire on time of urine sampling, illness in the past week, medication use on the day of urine collection, contact in the former 24 hours with: vehicle exhaust gasses, etheric oils/essences/candles, smoke, indoor heating sources, or cooking the evening before urine collection. The day before urine collection (Wednesday) participants filled out a food frequency questionnaire on food items possibly containing PAHs: coffee, cereals, muesli, bread, smoked fish or meat or cheese, chips, nuts, mayonnaise, cooking oil. Consumption in the 24h time-frame before urine collection was asked for, for the time periods before 8:00 in the morning, between 8:00-11:00, 11:00-14:00, 14:00-17:00, 17:00-20:00, 20:00-23:00, and after 23:00. Furthermore participants needed to record if they had been consuming barbecued or burnt food items in the day before urine collection.

*Table 1 Schematic overview of the PAH study protocol*

| **Sampling** | **Info** | **Menen** | **Genk/Diepenbeek** | **Houtem Background reference** |
|---|---|---|---|---|
| **AIR (24 PAHs)** | | | | |
| **Sampling locations** | near homes of people | 6 | 6 | 1 |
| **Sampling duration** | 48h consecutively | | | |
| **Sampling frequency** | times within one week | 2 | 2 | 2 |
| | number of weeks | 4 | 4 | 4 |
| | number of seasons | 2 | 2 | 2 |
| **FOOD** | | | | |
| **Sampling tool** | FFQ on past 24h | 20 | 20 | / |
| **Sampling frequency** | number of seasons | 2 | 2 | / |
| **URINE** | | | | |
| **Sampling tool** | First morning void | 20 | 20 | / |
| **Sampling frequency** | times within one week | 1 | 1 | / |
| | number of weeks | 4 | 4 | / |
| | number of seasons | 2 | 2 | / |

FFQ= food frequency questionnaire

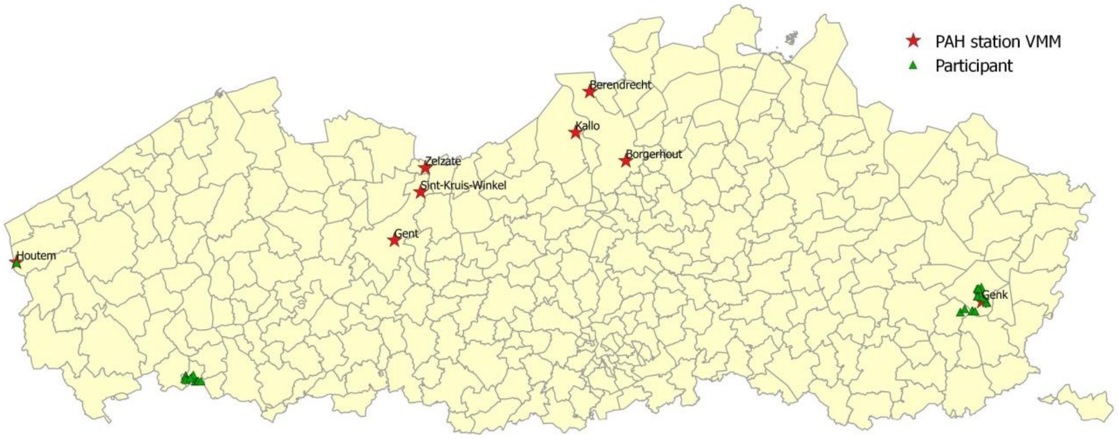

*Figure 2 Overview of all PAH stations of the measuring network of the Flemish Environmental Agency (VMM) (indicated by red star) and of all participants of the current study collecting PAHs and levoglucosan in the air in two seasons during 4 weeks each, and or repeatedly collecting urine in each of the sampling weeks (green triangle)*

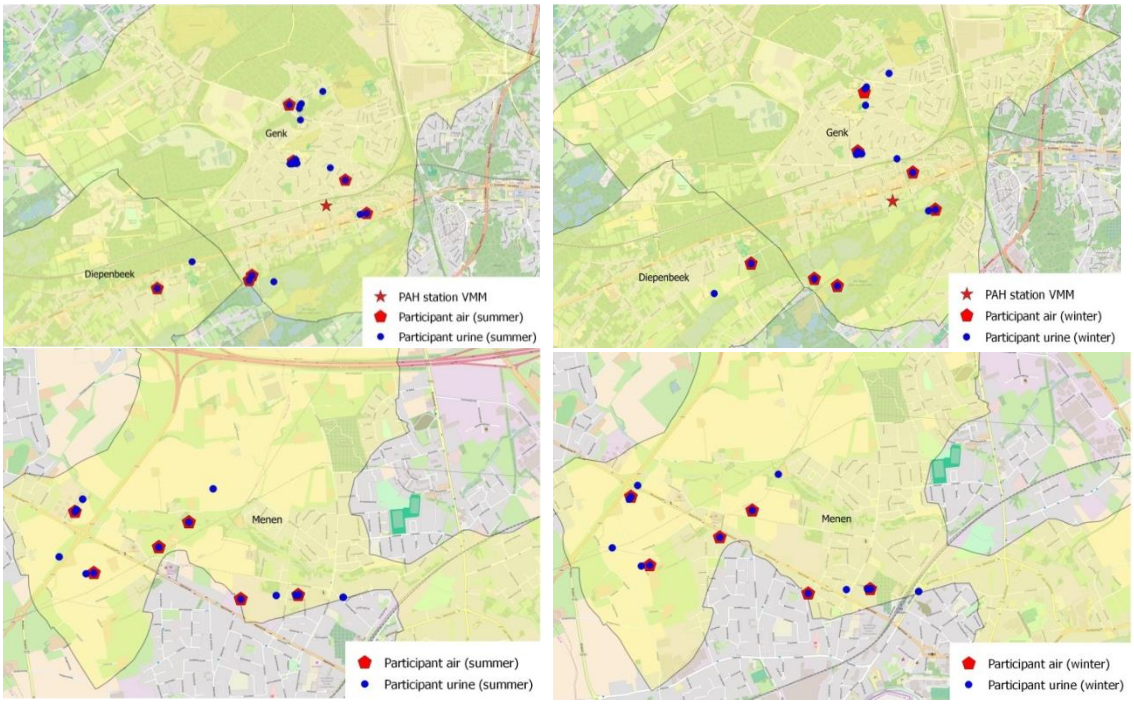

*Figure 3: Overview of participants in Genk/Diepenbeek (upper row) and in Menen (lower row) during the warm (left) and cold (right) season. Indication of the participants collecting air samples (red pentagon), and/or urine ( blue dots).*

## 2.4. PAH ANALYSIS

*Sampling cartridges*

The cartridges for sampling and thermal desorption were stainless-steel tubes (Markes International Ltd.) with the following dimensions: 31/2 inch length, ¼ inch o.d. and packed with PDMS/Tenax sorbent material. Prior to each use, the sampling tubes were conditioned by thermal cleaning under a nitrogen flow rate of 75 mL/min at 300°C for 60 min, sealed with end caps and stored under nitrogen atmosphere to prevent any contamination of the sorbent. After loading, the samples were immediately sealed again with the end caps and stored under nitrogen atmosphere until analysis. The samples were analyzed within seven days after the collection.

*Analysis*

In total 181 48h-air-samples were analyzed. Twenty-two different PAHs were measured in the air samples, namely the 16 EPA-PAHs, four methyl-PAHs (2-Me, and 1-Me-naphthalene, 9-Me-phenanthrene, 2-Me-anthracene), benzo(e)pyrene, and picene (Table 2). All the analyses were performed on a TD-GC-MS system, which consisted of a TD100 Thermal desorber (TD, Markes International Ltd.) and coupled to a gas chromatograph Thermo Trace GC Ultra and a mass selective detector Thermo DSQII (Thermo Fisher Scientific Inc.). Thermal desorption of the sampling tubes was carried out at 300°C with a flow rate of 20 mL/min for 12 min, followed by cold trap (10 °C) focusing. Thermal desorber system control was performed using Thermal Desorber System control Program version 4.4.1 (Markes International Ltd.). The GC-MS system control, data logging and data handling were performed using Xcalibur 2.0 software (Thermo Fisher Scientific Inc.). The chromatographic separation of the PAHs was conducted on Rxi-5Sil MS capillary column (Restek) 30m x 0.25mmi.d. x 0.25 mmfilm thickness. Helium was used as a carrier gas. All compounds were analyzed by using time scheduled Selected Ion Monitoring (SIM) of the most intensive ion fragment of each compound. The MS ion source and the transfer line from the GC to the MS were kept at 250 °C and 320°C respectively.

*Table 2 List of PAHs that are measured in the air samples. The so-called 16 EPA PAHs are indicated in green. Carcinogenicity probability according to IARC classification.*

| ID[a] | Abbreviation | Compounds | Number of rings | Molecular weight | IARC, vol.92, 2010 |
|---|---|---|---|---|---|
| **1** | NAP | naphthalene | 2 | 128.2 | 2B |
| **2** | 2MeNAP | 2-methylnaphthalene | 2 | 142.2 | - |
| **3** | 1MeNAP | 1-methylnaphthalene | 2 | 142.2 | - |
| **4** | ACY | acenaphthylene | 2 | 152.2 | - |
| **5** | ACE | acenaphthene | 2 | 154.2 | 3 |
| **6** | FLU | fluorene | 2 | 166.2 | 3 |
| **7** | PHE | phenanthrene | 3 | 178.2 | 3 |
| **8** | 9MePHE | 9-methylphenanthrene | 3 | 192.3 | - |
| **9** | ANT | anthracene | 3 | 178.2 | 3 |
| **10** | 2MeANT | 2-methyl anthracene | 3 | 192.3 | - |
| **11** | FLA | fluroanthene | 3 | 202.3 | 3 |
| **12** | PYR | pyrene | 4 | 202.1 | 3 |
| **13** | BaA | benz(a)anthracene | 4 | 228.3 | 2B |
| **14** | CHR | chrysene | 4 | 228.3 | 2B |
| **15** | BbF | benzo[b]fluoranthene | 4 | 252.3 | 2B |
| **16** | BkF | benzo[k]fluoranthene | 4 | 252.3 | 2B |
| **17** | BeP | benzo[e]pyrene | 5 | 252.3 | 3 |
| **18** | BaP | benzo[a]pyrene | 5 | 252.3 | 1 |
| **19** | IcdP | indeno(1,2,3-c,d)pyrene | 5 | 276.3 | 2B |
| **20** | DahA | dibenzo[a,h]anthracene | 5 | 278.3 | 2A |
| **21** | PIC | picene | 5 | 278.3 | 3 |

| ID[a] | Abbreviation | Compounds | Number of rings | Molecular weight | IARC, vol.92, 2010 |
|---|---|---|---|---|---|
| **22** | BghiP | benzo[g,h,i]perylene | 6 | 276.4 | 3 |

[a] *ID: identification number also used further on in some of the graphs of the report*

## 2.5. LEVOGLUCOSAN MEASUREMENTS IN THE OUTDOOR AIR

The levoglucosan (LEV) measurements were performed at TNO. A surface of 2.5 $cm^2$ of each quartz filter was used to measure levoglucosan. The analytical method for levoglucosan was described before by Simpson et al. (2004). Briefly, each filter was extracted in ethylacetate with 3.6 mM triethylamine in an ultrasonic bath for 1h. Further, samples were extracted with a silating reagent (TMSI). Levoglucosan was measured with gas chromatography in combination with mass spectrometric detection in electron impact mode (Agilent 6890/5973N GC/MS). Levoglucosan quantification is based on component identification by retention time, specific ion ratios and an internal standard (SRM2267). The expanded uncertainty (U) amounts 30%. Expanded uncertainty was calculated as 2 times the uncertainty (Uc) incorporating reproducibility (vc), recovery (utv) and accuracy of the calibration standard (uj), following the Dutch norm NEN 7777 Environment — Performance characteristics of measurement methods (Uc = √((vc)2 + (uj)2 + (utv)2)). Reproducibility of the method was between 7 and 15% depending on levoglucosan concentration in analyzed samples.

## 2.6. URINE MEASUREMENTS

1-OH pyrene, and urinary creatinine was measured in all 234 urine samples. The samples were collected once a week by all participants during each of the 4- weeks sampling period in both seasons.

Creatinine was analyzed using the Jaffe method in which creatinine precipitates after reaction with picric acid in an alkaline environment. 1-OH pyrene and OH-phenanthrenes were analyzed using Ultra Performance Liquid Chromatography (UPLC)-tandem MS. Urine samples were spiked with a mixture of ten $^{13}C$-labeled internal standards and sodium acetate buffer containing β-glucuronidase and sulfatase, hydrolyzed overnight at 37 °C, and then acetonitrile is added before injection. The instrumental analysis was performed by means of UPLC-tandem MS using a Waters H-class Acquity UPLC system (Waters, Milford, MA, USA). The UPLC system consisted of an Acquity quaternary solvent manager, an Acquity sample manager and an Acquity column heater manager. The PAHs metabolites were separated on an Acquity UPLC C18 column (100 mm × 2.1 mm; 1.7 µm). The column temperature was kept constant at 40°C. Optimum separation (gradient) was obtained with a binary mobile phase constituted of ultrapure water (solvent A) and acetonitrile (solvent B), both solvents acidified with 0.1% (v:v) acetic acid. The flow rate of the mobile phase was 0.4 mL/min. An aliquot of 100 µL of the urine sample was injected into the LC system. The UPLC system was coupled to a Waters Xevo TQ-S tandem mass spectrometer, which was operated in the negative electrospray ionization mode (ESI−). By recording the mass spectrum upon direct infusion of the compound into the mass spectrometer, both parameters were optimized in order to obtain maximum sensitivity with the highest amount of product ions available. Out of these results characteristic precursor and product ions were selected for detection in the multiple reaction monitoring (MRM) mode. For each compound the two highest precursor/product ion transitions are used. Positive identification of the PAHs metabolites were based on LC retention time match and their specific MRM transitions. Quantification of 1-hydroypyrene was done with the internal standard method by adding isotopically labelled analogues to the sample. The relative response factors (RRF) of the compounds in relation to the corresponding internal standard were calculated. Thus, the reported concentrations were corrected for recovery. The total area of the phenanthrene metabolites was obtained by summation of all areas of the different components (1-, 2-, 3-, 4- and 9-hydroxy-phenanthrene). The concentration was calculated qualitatively with the response factor of 1-hydroxypyrene (internal standard method), as native OH-phenanthrene reference standards, i.e. internal standards for the phenanthrene compounds were not used. Instead, SRM 3672 with certified values for 1-hydroxypyrene and the phenanthrene metabolites were used to estimate the response factor of the phenanthrene metabolites. The SRM 3672 was measured in every batch and the response factor per batch was determined. This response factor was used to calculate the levels of phenantrene metabolites (semi-quantitatively).

## 2.7. DATA ANALYSIS AND STATISTICS

The first hypothesis to test was: **Outdoor air PAH components and wood-burning tracer patterns will allow identification of specific PAH sources in Menen and Genk South**. Different techniques were used for source apportionment of PAHs: calculation of diagnostic ratios based on molecular tracers, and principal component analysis (PCA) and positive matrix factorization (PMF). A combination of these techniques are used as the application of one technique only can be insufficient to derive strong conclusions and a combination will reinforce the results (Gao et al., 2013). PCA is the most widely used tool, reducing the number of variables while retaining the original information as much as possible. Each PCA factor represents a profile that can either be associated with one source, multiple sources or conditions that effect the concentrations in a similar way (often weather conditions). PCA and PMF analysis were done using Statistica (Version 12 and the software of US-EPA (version 5.0.12), respectively. PMF is a multivariate factor analysis with non-negative constraints (no sample can have a negative source contribution). It decomposes a matrix of sample data into two matrices: factor contributions and factor profiles. It is often used on speciated $PM_{2.5}$ data sets with over 100 samples (Norris et al., 2008). Henry et al. (1984) suggested that the minimum adequate sample size (N) needed for obtaining statistically reliable results by PMF is $N= 30+(V + 3)/2$, where ‘V’ is the number of input species (compounds). In the current study respectively N=96, 70 and 15 valuable air samples were collected in Menen, Genk/Diepenbeek, and Houtem. In those samples 22 PAH compounds were measured, giving a sufficient amount to generate theoretically statistically reliable results using PMF on ‘regional’ (Menen or Genk/Diepenbeek) level, but a too low number of sampling points for PMF on ‘sampling location’ level.

The second hypothesis of the study was: **Outdoor air PAHs at different geographical locations, influence PAH exposure of people living near those locations**.

The following approach was followed to assess the correlation between markers in urine (1-OH pyrene and 1-OH phenantrene), and markers of external exposure (PAHs and the levoglucosan tracer). First, potential determinants of markers in urine were determined by mixed effects single linear regression. Mixed-effects models account for both fixed and random effects. As with all regression models, their purpose was to describe a response variable as a function of the predictor variables. Applying mixed-effects models, correlations within sample subgroups are taken into account. In this way, they provide a compromise between ignoring data groups entirely and fitting each group with a separate model. In this study, such correlations may be present at the level of participant-ID, because of the repeated measurement design. As such, ‘Participant ID’ was considered as random factor (random intercept). Urinary markers were adjusted for urinary creatinine levels and ln-transformed prior to analysis. The following variables were considered as potential determinant: season, region, past smoking, sex, family education, working outside of community, garage attached to house, age, BMI (both continuous and categorical), passive smoking, and some geographical parameters on habitation, land use and traffic. The subset of potential determinants for which the p-value of the association with urinary markers was less than 0.25 were selected to build a mixed effects multiple linear regression model. Following a stepwise approach, variables that were not significant at the 0.05 significance level were deleted from the model while starting with the least significant variable. The remaining subset was considered as covariates to adjust subsequent models assessing the association between PAH and levoglucosan exposure and urinary markers. PAH and the levoglucosan tracer exposure were $\log_2$ transformed prior to analysis. Using the 2-based logarithm enhances interpretation of the effect estimates, i.e. the estimated effect of a two-fold change in exposure. Again, a mixed effects multiple linear regression was applied, considering participant ID as random factor.

# CHAPTER 3 RESULTS

## 3.1. CHARACTERISTICS OF THE STUDY POPULATION AND THEIR LIFE ENVIRONMENT

In total 50 individuals participated in the study, 25 males and 25 females. The average age of the participants was 57.7 y, of which 51% were employed at diverse education levels (Table 3). All individuals were non-smokers, however 48% indicated ever having smoked. The average age of the participants' residences was 42 years (min 7 and max 120y). Almost half of the participants reported having a garage attached to their homes (48.0%). Most residences were heated with gas (70.0%) ,and had insulated roofs (87.5%). Of the in total 50 participants in the study, N=40 (77.5%) reported activities of industrial (N=16) and/or traffic (N=14) origin in the neighborhood of their house, causing increased exposure to combustion products. Some geographical parameters were calculated for the home environment of the participants. 13.9% of the residences were situated within a high residential area. They were either located in a busy road with more than 10000 vehicles/day or maximal ca. 1.2 km away from it (Table 3).

*Table 3: Characteristics of the in total 50 participants of the study considering host factors, life style, residence and home environment*

| Characteristics | | % or median (min-max) |
|---|---|---|
| **Host factors & life style** | | |
| % males | | 50.0 |
| Region: Genk-Diepenbeek / Menen | | 58 / 42 |
| age (y) | | 57.7 (33.3-75.8) |
| BMI (kg/m$^2$) | | 26.1 (19.4-39.9) |
| highest education level of family | no education | 4.1 |
| | primary school | 8.2 |
| | lower secondary level | 16.3 |
| | higher secondary level | 26.5 |
| | bachelor | 38.8 |
| | master | 6.1 |
| % single without partner | | 10.0 |
| % parents of Belgian origin | | 85.7 |
| % employed | | 51.0 |
| % of employed working outside the community | | 32.0 |
| % regularly in busy traffic (on weekdays and/or weekend) | | 24.5 |
| % former smoker | | 48.0 |

| Characteristics | | % or median (min-max) |
|---|---|---|
| number of years smoked | | 0.0 (0.0-36.0) |
| time in presence of smokers | | 0.0 (0.0-12.0) |
| % contact with vehicle exhaust in free time | | 12.8 |
| % contact with soot, ash in free time | | 4.8 |
| **Residence & home environment** | | |
| age of residence | | 42 (7-120) |
| habitable surface residence (m$^2$) | | 120 (65-250) |
| number of individuals in residence | | 2 (1-5) |
| type of residence | % detached residence | 24.0 |
| | % semi-detached residence | 36.0 |
| | % terraced residence/row house | 40.0 |
| % residences with insulated roof | | 87.5 |
| % residence heating on gas | | 70.0 |
| % residence heating on heating oil | | 26.0 |
| % using mothballs at home | | 8.2 |
| % attached garage to residence | | 48.0 |
| % car parking at home in asphalt | | 11.9 |
| % reporting activities in nearly residence region causing increased exposure to combustion | | 77.5 |
| road length in 50m buffer (m) | ROADLENGTH_50 | 166 (97-737) |
| traffic intensity on nearest road (veh/day) | Q_NEAR | 5944 (104-22857) |
| heavy traffic intensity on nearest road (veh/day) | Q_NEAR_HEAVY | 192 (0-2167) |
| traffic intensity x road length in 1000m buffer (veh*m/day*10$^6$) | TRAFLOAD_1000 | 46.0 (12.6-117.6) |
| heavy traffic intensity x road length in 1000m buffer (veh*m/day *10$^6$) | TRAFLOAD_1000_heavy | 2.7 (0.2-7.3) |
| Distance to major road (m) | 1/(D_NEAR_MAJOR1) | 334.4 (1.4-1245.7) |
| number of bus stops in 1000m buffer | BUSSTOPS_1000 | 22 (6-48) |
| high residential areas within 100m buffer? (Yes) [a] | HDRES_100 | 13.9 % |
| Surface of urban green within 3000m buffer (*10$^5$ m$^2$) [b] | UrbGr_3000 | 3.2 (0.0-7.5) |
| Surface of nature within 5000m buffer (*10$^6$m$^2$) [c] | NATURE_5000 | 42.2 (30.7-56.3) |

[a] high density residential areas of CORINE Class 111. [b] Urban green of CORINE class 141, 142. [c] Natural land of CORINE class 141, 142, 211, 231, 242, 311, 312, 313, 322, 421, 511, 522.

## 3.2. CHARACTERISTICS OF THE MEASURING LOCATIONS

PAHs and levoglucosan were measured on six measuring locations in each season in both Menen and Genk/Diepenbeek and one location in Houtem, near to residences. The residence types were summed up in Table 4. The measuring units of the traffic locations of Genk/Diepenbeek (G01, G21, G02 and G29) were located in streets with more than 10000 vehicles/day. Also on the locations GO3 (industry) and G06 (background) the nearest road (resp. 97 and 181 m away) was one with high traffic intensity. This was a bit different in Menen. The roads of the traffic locations, had much less (heavy) traffic (M05, M06). M05 differed from M06, as it had more passing traffic, whereas M06 showed rather peak hour traffic to the nearby industrial area. On the industry locations M01 and M11, there was still quite some traffic intensity in the neighborhood, whereas at location M02 there was much less traffic (only 104 vehicles per day on the nearest road, and the nearest major road (>10000 veh/day) being about 1.2 km away). At the sampled background residential locations of Menen (M03, M04), there was relatively high traffic intensity nearby (respectively 9686 and 5689 veh/day on roads respectively 139 and 173m away), and the nearest major road was about within a distance of 234 and 320m respectively. The surface of nature in a perimeter of 5 km was similar for all measuring locations (range of: 3.1 - 5.6E+07 $m^2$) (Table 5).

*Table 4: Measuring locations' characteristics*

| ID | location | Type of residence | Heating system | Sampling period |
|---|---|---|---|---|
| **Genk/Diepenbeek** | | | | |
| **G01** | traffic | terraced | gas | only summer-fall |
| **G21** | traffic | detached | gas | only winter (replacing G01) |
| **G02** | traffic | terraced | gas | only summer-fall |
| **G29** | traffic | semi-detached | gas | only winter (replacing G02) |
| **G03** | industry | detached | Pellet stove | both seasons |
| **G04** | industry | semi-detached | fuel | both seasons |
| **G05** | background | detached | gas | both seasons |
| **G06** | background | detached | gas | both seasons only levoglucosan sampling |
| **Menen** | | | | |
| **M01** | industry | detached | gas | only summer-fall |
| **M11** | industry | semi-detached | gas | only winter |
| **M02** | industry | semi-detached | gas | both seasons |
| **M03** | background | terraced | fuel | both seasons |
| **M04** | background | terraced | gas+electricity | both seasons |
| **M05** | traffic | terraced | gas | both seasons |
| **M06** | traffic | semi-detached | fuel+pellet stove | both seasons |
| **Houtem** | | | | |
| **H01** | background | terraced | Fuel+pellet/wood stove | both seasons |

*Table 5: Characteristics of the measuring location's environment*

| | G01 | G21 | G02 | G29 | G03 | G04 | G05 | G06 | M01 | M11 | M02 | M03 | M04 | M05 | M06 | H01 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **region** | **Genk/Diepenbeek** | | | | | | | | **Menen** | | | | | | | **Houtem** |
| **location type** | traffic | traffic | traffic | traffic | ind. | ind. | backgr. | backgr. | ind. | ind. | ind. | backgr. | backgr. | traffic | traffic | backgr. |
| **Sampling period** (1= summer/fall, 2= winter | 1 | 2 | 1 | 2 | 1+2 | 1+2 | 1+2 | 1+2 | 1 | 2 | 1+2 | 1+2 | 1+2 | 1+2 | 1+2 | 1+2 |
| **ROADLENGTH_50** (m) | 197.4 | 195.5 | 527.3 | 440.5 | 99.0 | 124.9 | 182.2 | 98.5 | 127.5 | 134.8 | 149.1 | 98.3 | 144.7 | 97.6 | 255.7 | 170.0 |
| **Q_NEAR** (veh/day) | 22348 | 22857 | 17173 | 11769 | 12307 | 4338 | 1477 | 13493 | 6422 | 6422 | 104 | 9686 | 5689 | 8918 | 6200 | 3941 |
| **DIST_NEAR (m)** | 12.7 | 21.6 | 1.4 | 20.5 | 96.8 | 45.1 | 70.3 | 180.9 | 295.5 | 312.3 | 341.8 | 139.4 | 172.7 | 1.0 | 38.2 | 1745.4 |
| **Q_NEAR_HEAVY** (veh/d) | 2167 | 2122 | 1054 | 0 | 1380 | 0 | 0 | 1573 | 134 | 134 | 0 | 178 | 208 | 564 | 742 | 30 |
| **1/(D_MAJOR ROAD1)** (m) | 12.7 | 21.6 | 1.4 | 20.5 | 96.8 | 1040.7 | 697.3 | 180.9 | 588.9 | 555.6 | 1175.0 | 234.3 | 320.1 | 117.5 | 477.8 | 4196.5 |
| **TRAFLOOAD_1000** (veh*m/d) | 6.1E+07 | 8.6E+07 | 1.2E+08 | 1.1E+08 | 4.7E+07 | 3.5E+07 | 4.2E+07 | 4.3E+07 | 3.7E+07 | 3.8E+07 | 2.1E+07 | 2.4E+07 | 5.0E+07 | 7.3E+07 | 4.8E+07 | 0 |
| **TRAFLOAD_1000_heavy** (veh*m/d) | 4.7E+06 | 7.4E+06 | 5.4E+06 | 4.3E+06 | 5.4E+06 | 5.6E+05 | 2.6E+06 | 3.2E+06 | 2.3E+06 | 2.3E+06 | 1.0E+06 | 8.9E+05 | 2.8E+06 | 4.3E+06 | 2.9E+06 | 0 |
| **BUSSTOPS_1000** | 37 | 25 | 47 | 47 | 12 | 23 | 14 | 20 | 17 | 18 | 18 | 19 | 22 | 32 | 28 | 2 |
| **HDRES_100** (0=neen/1=ja) | 0 | 0 | 1 | 1 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0 |
| **UrbGr_3000** (m$^2$) | 2.6E+05 | 6.4E+03 | 3.0E+05 | 3.2E+05 | 0.0E+00 | 3.2E+05 | 3.2E+05 | 3.2E+05 | 7.5E+05 | 7.5E+05 | 7.5E+05 | 7.5E+05 | 7.5E+05 | 7.5E+05 | 7.5E+05 | 0 |
| **NATURE_5000** (m$^2$) | 4.3E+07 | 3.8E+07 | 3.1E+07 | 3.1E+07 | 3.9E+07 | 3.1E+07 | 4.1E+07 | 3.7E+07 | 5.6E+07 | 5.6E+07 | 5.4E+07 | 5.1E+07 | 4.9E+07 | 5.4E+07 | 5.5E+07 | 5.3E+07 |

*With: ROADLENGTH_50= total road length in a 50m buffer around the residence, Q_NEAR = traffic intensity on nearest road, Q_NEAR_HEAVY: heavy traffic intensity on the nearest road , DIST_NEAR = distance to the nearest road where traffic counts were done (Q_NEAR), TRAFLOAD_1000 and TRAFLOAD_1000_HEAVY = traffic load being the multiplication of (all or only heavy) traffic intensity with road length in a perimeter of 1000m, 1/(D_NEAR_MAJOR1) = distance to the nearest major road (with > 10000 veh/day)) BUSSTOP_1000 = number of bus stops in perimeter of 1000m.*
*HDRES_100: high density residential areas (CORINE Class 111) in a perimeter of 100m, UrbGr_3000 = Urban green of CORINE class 141, 142 in a perimeter of 3000m, NATURE_5000 = natural land of CORINE class 141, 142, 211, 231, 242, 311, 312, 313, 322, 421, 511, 522 in a perimeter of 5000m*

## 3.3. METEOROLOGICAL CONDITIONS AND PARTICULATE MATTER LEVELS DURING THE SAMPLING PERIODS

The sampling was done in two seasons of 2015-2016 i.e. between August 31 and October 1, 2015, and between January 19 and March 5, 2016. The average temperatures in Sept'15 (13.6°C) and Jan-Feb'16 (5.2°C) were comparable with monthly normal temperatures for respectively the months September (14.9°C) and January-February (3.3 and 3.7°C) in the central measuring location of Ukkel (Belgium, period 1981-2010) (http://www.meteo.be/meteo/view/nl/360955-Maandelijkse+normalen.html#ppt_16230336). The median daily precipitation was in both seasons 0.20 mm i.e. 0.20 L/m$^2$. This was considerably lower than the normal values for the sampling months, being 4.38, 3.98 and 3.87 mm for September, January and February respectively.

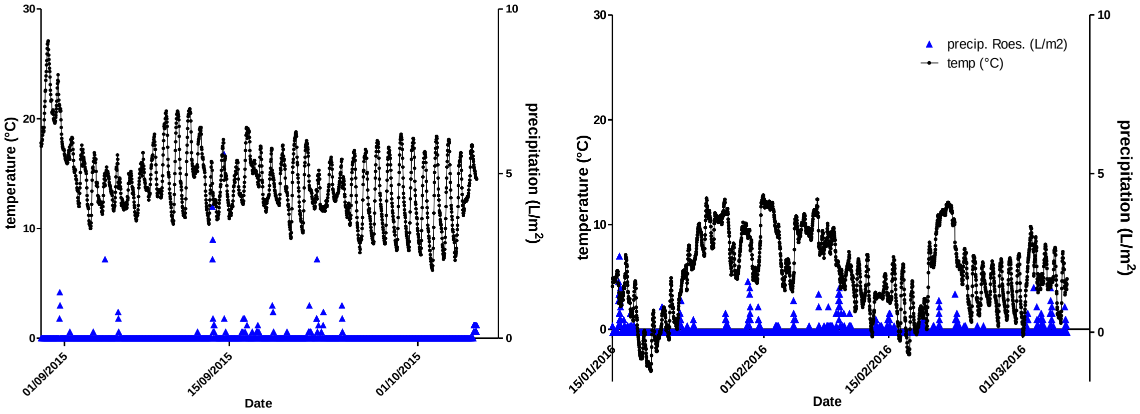

*Figure 4 Temperature and precipitation in the period of sampling (left: summer-fall, right: winter season)*

*Table 6: Meteorological conditions during the warm and cold sampling seasons (median(min-max) or % of the day). Calculated based on data collected by VMM.*

| Meteorological measurements | warm season | cold season |
|---|---|---|
| **Wind speed (m/s)** | 4.50 (0.25-13.29) | 6.59 (0.25-21.12) |
| **Temp (°C)** | 13.6 (4.9-22.7) | 5.2 (-4.1-12.4) |
| **Rel. humidity (%)** | 90 (44-100) | 90 (56-100) |
| **Precipitation per day (mm = L/m$^2$)** | 0.20 (0.00-20.20) | 0.20 (0.00-8.60) |
| **wind from West direction (SW, W, NW)** | 54.0% of the days | 60.0% of the days |
| **wind from East direction (NE, E, SE)** | 36.4% of the days | 16.7% of the days |

During both sampling periods, the levels of PM$_{10}$ and PM$_{2.5}$ were relatively low. The median level in the summer-fall and winter sampling periods (over three sampling locations, Figure 5) were respectively for PM$_{10}$ 14.0 and 17.0 µg/m$^3$, and for PM$_{2.5}$ 7.0 and 10.0 µg/m$^3$. At the start of the winter period (end of January 2016) there was however a short smog period. In that week there were respectively two (Menen) and one day(s) (Hasselt) with PM$_{10}$ levels above the EU 24h guideline value of 50µg/m$^3$. In the second halve of February there were also some days with moderately higher PM levels. Those periods occurred during some colder days in January and February.

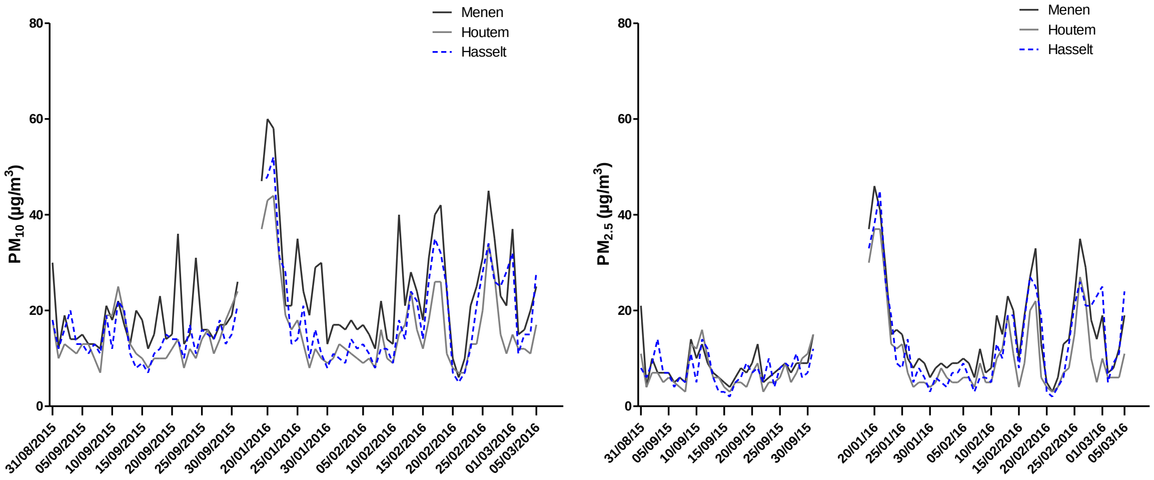

*Figure 5 PM$_{10}$ (left) and PM$_{2.5}$ (right) 24h-averaged levels at three measuring units of the Flemish Environmental Agency VMM within or near to the sampling areas of the current study, namely: Menen (Wervikstraat, id. code: 40MN01), Houtem (Westmoerstraat, id. code: 44N029), and Hasselt (Boksbeemdenstraat, id. code: 42N045). The latter station being the nearest PM-station to Genk.*

## 3.4. CONCENTRATION OF PAHS AND LEVOGLUCOSAN IN THE DIFFERENT SAMPLING AREAS

### 3.4.1. OVERALL COMPARISON MENEN, GENK, HOUTEM

A first overall comparison of the different sampling areas, showed that in summer-fall (Sept'15), the level of PAHs and levoglucosan measured in Menen and Genk/Diepenbeek were higher than those in the reference background station Houtem. This was not observed in the winter campaign (Jan-Feb'16). In that season the volatile lower molecular weight PAHs were generally not significantly different among the regions, and some 4-, 5- and 6-ring PAHs were higher in the background reference Houtem compared to the other regions. Further comparison was done on the level of the measuring units as this gives better insight in the local differences (see: 3.4.2 and 3.4.7).

*Table 7: Comparison of the levels of PAHs (ng/m$^3$) among the different regions. Results of Mann-Withney U-test for each of the compounds (p-values for comparison of Genk vs. Menen, Genk vs. Houtem, and Menen vs.Houtem)*

| **compound** | **Sept'15** | | | | | | | | | **Jan-Feb'16** | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **Genk** | | **Menen** | | **Houtem** | | **p-value** | | | **Genk** | | **Menen** | | **Houtem** | | **p-value** | | |
| | **N** | **Med** | **N** | **Med** | **N** | **Med** | **G-M** | **G-H** | **M-H** | **N** | **Med** | **N** | **Med** | **N** | **Med** | **G-M** | **G-H** | **M-H** |
| **NAP** | 39 | 12.99 | 42 | 20.25 | 7 | 1.77 | 0.027 | 0.000 | 0.000 | 31 | 21.90 | 41 | 16.71 | 8 | 30.09 | 0.847 | 0.328 | 0.394 |
| **2MeNAP** | 39 | 3.20 | 42 | 10.87 | 7 | 0.60 | 0.005 | 0.001 | 0.000 | 31 | 5.04 | 41 | 4.73 | 8 | 6.24 | 0.617 | 0.597 | 0.776 |
| **1MeNAP** | 39 | 1.71 | 42 | 6.18 | 7 | 0.22 | 0.002 | 0.003 | 0.000 | 31 | 3.47 | 41 | 3.08 | 8 | 4.02 | 0.502 | 0.550 | 0.715 |
| **ACY** | 39 | 0.47 | 42 | 0.15 | 7 | 0.01 | 0.006 | 0.000 | 0.001 | 31 | 0.17 | 41 | 0.14 | 8 | 0.17 | 0.856 | 0.772 | 0.735 |
| **ACE** | 39 | 1.63 | 42 | 1.24 | 7 | 0.01 | 0.086 | 0.000 | 0.000 | 31 | 0.41 | 41 | 0.49 | 8 | 0.81 | 0.794 | 0.695 | 0.164 |
| **FLU** | 39 | 0.84 | 42 | 2.30 | 7 | 0.06 | 0.092 | 0.008 | 0.000 | 31 | 0.61 | 41 | 0.87 | 8 | 1.70 | 0.724 | 0.462 | 0.108 |
| **PHE** | 39 | 2.61 | 42 | 4.60 | 7 | 0.11 | 0.251 | 0.000 | 0.000 | 31 | 2.25 | 41 | 2.30 | 8 | 4.38 | 0.707 | 0.597 | 0.190 |
| **9MePHE** | 39 | 0.52 | 42 | 0.42 | 7 | 0.01 | 0.018 | 0.001 | 0.004 | 31 | 0.08 | 41 | 0.09 | 8 | 0.09 | 0.280 | 0.462 | 0.598 |
| **ANT** | 39 | 0.16 | 42 | 0.01 | 7 | 0.01 | 0.000 | 0.001 | 0.415 | 31 | 0.10 | 41 | 0.15 | 8 | 0.24 | 0.794 | 0.222 | 0.081 |
| **2MeANT** | 39 | 0.25 | 42 | 0.22 | 7 | 0.01 | 0.204 | 0.000 | 0.000 | 31 | 0.08 | 41 | 0.02 | 8 | 0.06 | 0.088 | 0.645 | 0.449 |
| **FLA** | 39 | 0.90 | 42 | 0.85 | 7 | 0.01 | 0.536 | 0.000 | 0.000 | 31 | 0.85 | 41 | 0.57 | 8 | 1.40 | 0.521 | 0.550 | 0.172 |
| **PYR** | 39 | 0.80 | 42 | 0.66 | 7 | 0.01 | 0.163 | 0.000 | 0.000 | 31 | 0.50 | 41 | 0.38 | 8 | 0.91 | 0.682 | 0.505 | 0.181 |
| **BaA** | 39 | 0.87 | 42 | 0.44 | 7 | 0.04 | 0.000 | 0.000 | 0.000 | 31 | 0.21 | 41 | 0.17 | 8 | 0.40 | 0.617 | 0.085 | 0.027 |

| **compound** | **Sept'15** | | | | | | | | | | | **Jan-Feb'16** | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **Genk** | | **Menen** | | **Houtem** | | **p-value** | | | | | **Genk** | | **Menen** | | **Houtem** | | **p-value** | | |
| | **N** | **Med** | **N** | **Med** | **N** | **Med** | **G-M** | **G-H** | **M-H** | **N** | **Med** | **N** | **Med** | **N** | **Med** | **G-M** | **G-H** | **M-H** |
| **CHR** | 39 | 0.53 | 42 | 0.38 | 7 | 0.03 | 0.000 | 0.000 | 0.000 | 31 | 0.22 | 41 | 0.14 | 8 | 0.44 | 0.109 | 0.621 | 0.096 |
| **BbF** | 39 | 0.81 | 42 | 0.57 | 7 | 0.04 | 0.000 | 0.000 | 0.001 | 31 | 0.31 | 41 | 0.12 | 8 | 0.53 | 0.004 | 0.279 | 0.000 |
| **BkF** | 39 | 0.53 | 42 | 0.25 | 7 | 0.01 | 0.000 | 0.000 | 0.004 | 31 | 0.18 | 41 | 0.01 | 8 | 0.11 | 0.000 | 0.073 | 0.018 |
| **BeP** | 39 | 0.64 | 42 | 0.31 | 7 | 0.01 | 0.000 | 0.000 | 0.000 | 31 | 0.19 | 41 | 0.01 | 8 | 0.14 | 0.000 | 0.264 | 0.000 |
| **BaP** | 39 | 0.76 | 42 | 0.34 | 7 | 0.02 | 0.000 | 0.000 | 0.000 | 31 | 0.22 | 41 | 0.11 | 8 | 0.31 | 0.000 | 0.959 | 0.000 |
| **IcdP** | 39 | 0.95 | 42 | 0.27 | 7 | 0.10 | 0.000 | 0.000 | 0.002 | 31 | 0.16 | 41 | 0.18 | 8 | 0.19 | 0.369 | 0.695 | 0.015 |
| **DahA** | 39 | 1.02 | 42 | 0.27 | 7 | 0.01 | 0.000 | 0.000 | 0.004 | 31 | 0.15 | 41 | 0.20 | 8 | 0.22 | 0.002 | 0.048 | 0.004 |
| **PIC** | 39 | 1.02 | 42 | 0.18 | 7 | 0.01 | 0.000 | 0.000 | 0.175 | 31 | 0.11 | 41 | 0.32 | 8 | 0.35 | 0.000 | 0.037 | 0.011 |
| **BghiP** | 39 | 0.82 | 42 | 0.17 | 7 | 0.10 | 0.000 | 0.000 | 0.054 | 31 | 0.13 | 41 | 0.17 | 8 | 0.18 | 0.027 | 0.123 | 0.015 |
| **Sum 22PAHs** | **39** | **45.0** | **42** | **50.1** | **7** | **3.60** | 0.567 | 0.000 | 0.000 | **31** | **42.2** | **41** | **31.0** | **8** | **54.3** | 0.999 | 0.401 | 0.409 |
| **LEV** | **33** | **33.2** | **42** | **44.6** | **22** | **24.3** | 0.072 | 0.075 | 0.001 | **46** | **118.5** | **48** | **210.0** | **30** | **87.7** | 0.011 | 0.070 | 0.001 |

G=Genk, M=Menen, H=Houtem; LEV=levoglucosan

### 3.4.2. SEASONAL VARIATION OF PAH AND LEV LEVELS ON THE INDIVIDUAL SAMPLING STATIONS

For the monitoring stations that were sampled in both seasons, the ratios of the average PAH and LEV concentrations measured during winter (Feb-Jan'16) over summer-fall (Sept'15) were calculated (Table 8). At all locations, the levels of levoglucosan were higher (factor 2-7) in the winter compared to the September campaign. The same was true for the atmospheric PAHs measured at the reference (background) monitoring station of Houtem: average ratios from 2 (9MePHE) to 96 (ACE) were observed. This was in contrast with the background monitoring stations of Genk (G05) and Menen (M03 and M04) that had higher concentrations for almost all PAHs in September compared to the winter time. The 'industry' monitoring stations[5] (Genk stations G03 and G04 and Menen station M02) had generally higher concentrations of the relatively low molecular weight PAHs during winter compared to the summer-fall, whereas the opposite was true for (some of) the relatively high molecular weight PAHs. Regarding the two traffic monitoring stations of Menen (M05 and M06), no trend could be noticed: station M05 had higher winter levels of IcdP, BahA and BghiP while station M06 showed higher winter levels for some lower molecular weight PAHs as well.

The ratio of BaP over the sum of BaP and BeP was calculated to assess photosensitivity of PAHs. BaP is photodegraded more rapidly than its isomer BeP. If this ratio is below 0.5, it is often considered that there is atmospheric ageing of gaseous and particle PAHs (Tobiszewski, 2012). During the four weeks sampling period in summer-fall and winter this ratio was stable for Genk (average 0.53 (stdev=0.05), and 0.59 (0.11) respectively) and Menen (0.53 (0.06), and 0.92 (0.07) respectively). This means that photodecomposition of PAHs could be considered to be marginal.

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[5] Definition see 2.1

*Table 8 Average PAH and LEV concentrations for the monitoring stations which had repeated sampling during summer and winter (M02, M03, M04, M05, G03, G04, G05 and H01). *: ratio of the average PAH or LEV concentration measured during winter (Jan-Feb'16) over summer-fall (Sept'15); values higher or lower than 1 are indicated in blue (conc. higher in winter), and red (conc. higher in summer-fall).*

| conc (ng/m3) | | | NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP | LEV |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Menen | M02 | Avg winter | 30.631 | 8.621 | 5.702 | 0.784 | 0.811 | 1.590 | 4.645 | 0.126 | 0.426 | 0.130 | 1.299 | 0.980 | 0.324 | 0.430 | 0.304 | 0.032 | 0.045 | 0.163 | 0.125 | 0.192 | 0.313 | 0.125 | 377 |
| | industry | Avg summer | 15.436 | 6.360 | 3.535 | 0.143 | 0.804 | 1.352 | 2.675 | 0.265 | 0.040 | 0.196 | 0.604 | 0.489 | 0.463 | 0.384 | 0.536 | 0.327 | 0.377 | 0.438 | 0.444 | 0.390 | 0.341 | 0.248 | 68 |
| | | Ratio* | 2.0 | 1.4 | 1.6 | 5.5 | 1.0 | 1.2 | 1.7 | 0.5 | 10.6 | 0.7 | 2.1 | 2.0 | 0.7 | 1.1 | 0.6 | 0.1 | 0.1 | 0.4 | 0.3 | 0.5 | 0.9 | 0.5 | 5.535 |
| | M03 | Avg winter | 24.809 | 6.792 | 4.838 | 0.194 | 0.573 | 1.198 | 2.996 | 0.086 | 0.203 | 0.049 | 0.800 | 0.552 | 0.257 | 0.271 | 0.212 | 0.028 | 0.010 | 0.128 | 0.125 | 0.198 | 0.321 | 0.125 | 307 |
| | background | Avg summer | 33.552 | 15.374 | 10.127 | 0.366 | 1.805 | 3.399 | 6.840 | 0.337 | 0.162 | 0.354 | 1.166 | 1.117 | 0.449 | 0.361 | 0.499 | 0.136 | 0.260 | 0.265 | 0.233 | 0.178 | 0.275 | 0.262 | 51 |
| | | Ratio* | 0.7 | 0.4 | 0.5 | 0.5 | 0.3 | 0.4 | 0.4 | 0.3 | 1.3 | 0.1 | 0.7 | 0.5 | 0.6 | 0.8 | 0.4 | 0.2 | 0.0 | 0.5 | 0.5 | 1.1 | 1.2 | 0.5 | 6.065 |
| | M04 | Avg winter | 19.881 | 5.418 | 3.530 | 0.227 | 0.442 | 0.868 | 2.557 | 0.088 | 0.177 | 0.043 | 0.725 | 0.524 | 0.226 | 0.194 | 0.130 | 0.009 | 0.005 | 0.100 | 0.125 | 0.196 | 0.318 | 0.125 | 348 |
| | background | Avg summer | 29.300 | 10.347 | 6.568 | 0.153 | 1.162 | 2.325 | 4.765 | 0.268 | 0.005 | 0.268 | 0.854 | 0.743 | 0.313 | 0.238 | 0.343 | 0.065 | 0.124 | 0.167 | 0.143 | 0.191 | 0.150 | 0.146 | 89 |
| | | Ratio* | 0.7 | 0.5 | 0.5 | 1.5 | 0.4 | 0.4 | 0.5 | 0.3 | 35.4 | 0.2 | 0.8 | 0.7 | 0.7 | 0.8 | 0.4 | 0.1 | 0.0 | 0.6 | 0.9 | 1.0 | 2.1 | 0.9 | 3.908 |
| | M05 | Avg winter | 13.497 | 3.944 | 2.501 | 0.131 | 0.430 | 0.567 | 1.754 | 0.089 | 0.082 | 0.029 | 0.596 | 0.386 | 0.268 | 0.227 | 0.097 | 0.005 | 0.005 | 0.096 | 0.125 | 0.202 | 0.329 | 0.125 | 293 |
| | traffic | Avg summer | 40.001 | 16.266 | 10.675 | 0.420 | 3.720 | 4.457 | 8.955 | 0.483 | 0.358 | 0.423 | 1.631 | 1.209 | 0.482 | 0.464 | 0.603 | 0.074 | 0.369 | 0.340 | 0.125 | 0.168 | 0.344 | 0.125 | 59 |
| | | Ratio* | 0.3 | 0.2 | 0.2 | 0.3 | 0.1 | 0.1 | 0.2 | 0.2 | 0.2 | 0.1 | 0.4 | 0.3 | 0.6 | 0.5 | 0.2 | 0.1 | 0.0 | 0.3 | 1.0 | 1.2 | 1.0 | 1.0 | 4.978 |
| Genk | G03 | Avg winter | 46.357 | 11.706 | 7.601 | 0.283 | 1.846 | 3.234 | 8.542 | 0.135 | 0.584 | 0.227 | 2.396 | 1.636 | 0.537 | 0.751 | 0.808 | 0.463 | 0.513 | 0.535 | 0.359 | 0.285 | 0.295 | 0.228 | 146 |
| | industry | Avg summer | 15.450 | 4.469 | 2.290 | 0.142 | 0.910 | 1.290 | 3.512 | 0.487 | 0.178 | 0.252 | 0.929 | 0.874 | 0.739 | 0.419 | 0.687 | 0.385 | 0.445 | 0.531 | 0.595 | 0.602 | 0.384 | 0.434 | 36 |
| | | Ratio* | 3.0 | 2.6 | 3.3 | 2.0 | 2.0 | 2.5 | 2.4 | 0.3 | 3.3 | 0.9 | 2.6 | 1.9 | 0.7 | 1.8 | 1.2 | 1.2 | 1.2 | 1.0 | 0.6 | 0.5 | 0.8 | 0.5 | 4.078 |
| | G04 | Avg winter | 8.343 | 1.732 | 1.155 | 0.382 | 0.141 | 0.271 | 0.935 | 0.064 | 0.047 | 0.028 | 0.338 | 0.238 | 0.171 | 0.181 | 0.188 | 0.117 | 0.151 | 0.201 | 0.218 | 0.190 | 0.206 | 0.188 | 183 |
| | industry | Avg summer | 9.290 | 1.345 | 0.516 | 1.216 | 8.828 | 0.070 | 0.364 | 0.151 | 0.135 | 0.182 | 0.299 | 0.297 | 0.617 | 0.537 | 0.737 | 0.587 | 0.638 | 0.821 | 1.017 | 1.197 | 1.159 | 0.808 | 43 |
| | | Ratio* | 0.9 | 1.3 | 2.2 | 0.3 | 0.0 | 3.9 | 2.6 | 0.4 | 0.3 | 0.2 | 1.1 | 0.8 | 0.3 | 0.3 | 0.3 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 4.209 |
| | G05 | Avg winter | 10.583 | 2.760 | 1.762 | 0.123 | 0.475 | 0.720 | 2.333 | 0.074 | 0.163 | 0.080 | 0.728 | 0.454 | 0.228 | 0.341 | 0.299 | 0.156 | 0.192 | 0.237 | 0.222 | 0.200 | 0.227 | 0.189 | 130 |
| | background | Avg summer | 15.235 | 3.849 | 2.200 | 0.253 | 0.821 | 1.201 | 3.367 | 0.313 | 0.053 | 0.210 | 0.691 | 0.559 | 0.586 | 0.435 | 0.676 | 0.456 | 0.517 | 0.614 | 0.715 | 0.877 | 0.993 | 0.424 | 51 |
| | | Ratio* | 0.7 | 0.7 | 0.8 | 0.5 | 0.6 | 0.6 | 0.7 | 0.2 | 3.1 | 0.4 | 1.1 | 0.8 | 0.4 | 0.8 | 0.4 | 0.3 | 0.4 | 0.4 | 0.3 | 0.2 | 0.2 | 0.4 | 2.566 |
| Houtem | H | Avg winter | 27.838 | 6.201 | 4.155 | 0.174 | 0.792 | 1.596 | 4.061 | 0.099 | 0.283 | 0.073 | 1.234 | 0.833 | 0.365 | 0.418 | 0.490 | 0.098 | 0.138 | 0.276 | 0.125 | 0.226 | 0.348 | 0.125 | 190 |
| | background | Avg summer | 2.544 | 0.666 | 0.299 | 0.005 | 0.009 | 0.096 | 0.282 | 0.048 | 0.009 | 0.015 | 0.018 | 0.022 | 0.062 | 0.038 | 0.105 | 0.013 | 0.011 | 0.057 | 0.125 | 0.018 | 0.028 | 0.125 | 27 |
| | | Ratio* | 10.9 | 9.3 | 13.9 | 34.7 | 84.5 | 16.6 | 14.4 | 2.0 | 31.7 | 5.0 | 70.4 | 38.0 | 5.9 | 11.1 | 4.7 | 7.8 | 12.3 | 4.8 | 1.0 | 12.4 | 12.7 | 1.0 | 6.942 |

### 3.4.3. COMPARISON WITH AVAILABLE VMM DATA

For Genk and Houtem, a comparison could be made between the current results of summer-fall (Sept'15) PAH measurements performed in those areas, and data of the Flemish environmental Agency (VMM) collected in the same period. It should however be noted that the PAHs were sampled and measured with different techniques: In the current study, cartridges were used, trapping both the gas and particle phase. VMM is obliged to use the EU reference method and therefore sampled the $PM_{10}$ particle phase via filters allowing only analysis of the higher molecular weight PAHs (Table 9). Wauters et al. (2008) reported that average PAH concentrations sampled via sorbent tube (and measured via GC-MS) were systematically higher than those collected with high-volume samplers (extracted with ASE and analysed via HPLC with fluorescence detection). For the higher MW compounds the difference (ratio of concentration measured on tubes versus concentration measured by high-volume sampling) was in the order of a factor 1.2–3. For the more volatile naphthalene and acenaphthene, the concentrations measured on the sorbent tube were respectively 35 and 23 times higher. Losses probably occur due to sample preparation (Soxhlet extraction or ASE, concentration), together with a temperature effect during sampling and possible chemical reactions with inorganic gases like ozone, NO*x*, and $SO_2$ on the sampling filters. Wauters et al. (2008) indeed showed that the differences between the two methods were larger in summer than in winter. For the low molecular weight PAHs, the large difference between the values obtained by the method using the mixed bed trap in combination with thermal desorption GC–MS was mainly explained by losses by the high-volume sampling due to blow-off effects caused by the high air velocity through the filter.  Furthermore, the sampling dates were not exactly the same (24h averages for VMM and 48h averages in the current study), which might influence the levels, as PAH concentrations may fluctuate considerable from day to day.

Aside from these technical aspect, the VMM monitoring station in Genk could be classified as a background station, while those in the current study were situated at a background, two traffic, and two industry locations. Indeed, on average relatively high PAH concentrations (for all eight considered PAHs) were measured in the five stations of Genk of the current study, compared to the median VMM results in Genk. In Houtem, the in the current study measured levels were all in the same range than the VMM levels measured during the same days. The Houtem measuring units of both studies were located close to each other at a distance of less than 100m.

*Table 9 Median(min-max) PAH levels measured in the current study compared to those of the VMM monitoring campaign in the summer-fall period (30/08/15-2/10/15). Important to note is that different sampling and analysis techniques were used in both studies (see text)*

| PAH | Genk | | Houtem | |
|---|---|---|---|---|
| **(ng/$m^3$)** | **VMM(1)** | **Current study(2)** | **VMM(1)** | **Current study(2)** |
| **FLA** | 0.11 (0.05-0.37) | 0.84 (0.01-11.24) | 0.05 (0.02-0.19) | 0.06 (0.01-0.27) |
| **PYR** | 0.11 (0.04-0.31) | 0.80 (0.01-7.75) | 0.04 (0.01-0.16) | 0.01 (0.01-0.05) |
| **BaA** | 0.05(0.02-0.17) | 0.87 (0.01-1.24) | 0.02 (0.001-0.05) | 0.04 (0.01-0.18) |
| **CHR** | 0.13 (0.06-0.37) | 0.53 (0.01-3.47) | 0.05 (0.01-0.19) | 0.03 (0.01-0.08) |
| **BbF** | 0.16 (0.06-0.43) | 0.81 (0.01-5.28) | 0.04 (0.01-0.18) | 0.04 (0.01-0.31) |
| **BkF** | 0.08 (0.03-0.22) | 0.53 (0.01-5.57) | 0.02 (0.003-0.09) | 0.01 (0.01-0.04) |
| **BaP** | 0.10 (0.03-0.19) | 0.76 (0.01-6.99) | 0.03 (0.01-0.08) | 0.02 (0.01-0.14) |
| **IcdP** | 0.13 (0.05-0.24) | 0.95 (0.01-11.47) | 0.02 (0.001-0.12) | 0.01 (0.01-0.05) |
| **DahA** | 0.02 (0.01-0.05) | 1.02 (0.01-14.71) | 0.01 (0.001-0.02) | 0.01 (0.01-0.06) |
| **BghiP** | 0.16 (0.06-0.31) | 0.81 (0.01-9.32) | 0.02 (0.01-0.14) | 0.01 (0.01-0.07) |

(1) Median (minimum-maximum) of VMM measurements in summer-fall sampling period of the current study

(2) Median (minimum-maximum) of 48-h concentrations from summer-fall sampling period, measured at different monitoring stations within that area

The winter levoglucosan concentrations of the current study could be compared with those in a recent VMM wood burning project. In the current study levoglucosan was measured in $PM_{2.5}$, whereas in the VMM study $PM_{10}$ was sampled for the analysis. The levoglucosan levels, measured on the overlapping days in the second half of January '16, were high in both studies (up to more than 900 $ng/m^3$). It is interesting to observe that there was a similar trend in levoglucosan measurements on all measuring locations on the same sampling days. Also on the VITO ('M1') location, which is a company site without wood burning sources, high levoglucosan levels were observed on January 19 and 21 (Table 11). This might be due to longer-range aerosol transport leading to rather overall concentrations at different measuring locations (levoglucosan is a stable compound).

Multiplying the levogluosan concentration with a factor of 10.7 one can calculate the $PM_{10}$ or $PM_{2.5}$ woodsmoke mass (Schmidl et al., 2008). That factor has been derived from emission testing in Austria and may be different elsewhere in Europe. The levoglucosan content of wood smoke depends on the type of wood burnt (soft or hard wood), temperature and type of burning process. The estimated uncertainty in particulate matter levoglucosan content has been estimated to be about 30% (Maenhaut et al., 2012). For each of the measurements on all sampling locations, the $PM_{2.5}$ levoglucosan concentration was multiplied by 10.7, to calculate the local contribution of wood burning to the $PM_{2.5}$ levels (measured by VMM on nearby sampling units). In summer-fall, as well as in winter, the contributions were relatively high (median levels less than 10% to up to 20.3%). These percentages were also observed during a levoglucosan measuring campaign performed in the year 2010 on several locations in Flanders (Maenhaut et al., 2012) (Table 10). Recently Maenhaut et al. (2016) reported a for Flanders adapted biomass PM contribution factor of 22,6. Applying that factor to our data, the estimated contribution of biomass burning to $PM_{2.5}$ would be between lower than ca. 10% and about 40%.

*Table 10: Percentage of $PM_{2.5}$ mass originating from wood smoke. $PM_{2.5}$ was measured on nearby VMM measuring stations. For the current study, the calculation of % wood burning contribution to $PM_{2.5}$ was done for each of the sampling days, and then the median($P_{25}$-$P_{75}$) of each region was calculated. From the study of Maenhaut et al.(2012), the lowest and highest average for each of the 7 different measuring locations are given*

| **Season** | | **Current study** | | **Study of Maenhaut et al. (2012)** |
|---|---|---|---|---|
| | **Genk** | **Menen** | **Houtem** | **range of averages on 7 measuring units** |
| **Summer** | 8.1 (4.9-18.0) | 8.4 (5.9-12.5) | 5.0 (2.9-6.9) | 0.5-5.0 |
| **Autumn** | | | | 6.5-18.9 |
| **Winter** | 14.9 (10.6-21.3) | 20.3 (14.1-34.2) | 16.2 (12.5-19.8) | 9.2-21.9 |

On January 19 and 21 the average daily temperature was around or just above freezing point, and the $PM_{10}$ and $PM_{2.5}$ levels were high (Table 1). On those days the contribution of wood burning to $PM_{10}$ (VMM study) or $PM_{2.5}$ (current study) was between 7 and 30% in Dessel, Mol, Genk and Houtem. On January 24 and 26, the levoglucosan levels and also $PM_{10}$ and $PM_{2.5}$ dropped significantly, giving a mass % $PM_{10}$ or $PM_{2.5}$ contribution from wood burning, of 15-26% on Jan24 and 2-13% on Jan26 (Table 11).

*Table 11. Comparison of air levoglucosan concentrations measured in the current study and the VMM wood burning study. The % of $PM_{10}$ originating from wood smoke is calculated ('wood')*

| Date ( av. day temp) | | 19jan'16 (0°C) | | | | 21 jan'16 (2.4°C) | | | | 24 jan'16 (9.4°C) | | | | 26 jan'16 (10°C) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | LEV[a] ng/m$^3$ | $PM_{10}$[b] µg/m$^3$ | $PM_{2.5}$[b] µg/m$^3$ | wood % | LEV[a] ng/m$^3$ | $PM_{10}$[b] µg/m$^3$ | $PM_{2.5}$[b] µg/m$^3$ | wood % | LEV[a] ng/m$^3$ | $PM_{10}$[b] µg/m$^3$ | $PM_{2.5}$[b] µg/m$^3$ | wood % | LEV[a] ng/m$^3$ | $PM_{10}$[b] µg/m$^3$ | $PM_{2.5}$[b] µg/m$^3$ | wood % |
| **VMM study** | **D1** | 715 | 46 | 33 | 17 | 752 | 52 | 45 | 15 | 182 | 13 | 9 | 15 | - | 21 | 14 | - |
| | **D2** | 808 | 46 | 33 | 19 | 769 | 52 | 45 | 16 | 184 | 13 | 9 | 15 | - | 21 | 14 | - |
| | **D3** | 761 | 46 | 33 | 18 | 804 | 52 | 45 | 17 | 282 | 13 | 9 | 23 | - | 21 | 14 | - |
| | **M1** | 695 | 46 | 33 | 16 | 897 | 52 | 45 | 18 | 162 | 13 | 9 | 13 | - | 21 | 14 | - |
| **Current study** | **G21** | 938 | 46 | 33 | 30 | - | 52 | 45 | - | 219 | 13 | 9 | 26 | - | 21 | 14 | - |
| | **G29** | 769 | 46 | 33 | 25 | 444 | 52 | 45 | 11 | - | 13 | 9 | - | 101 | 21 | 14 | 8 |
| | **G03** | - | 46 | 33 | - | - | 52 | 45 | - | - | 13 | 9 | - | 85 | 21 | 14 | 6 |
| | **G04** | 620 | 46 | 33 | 20 | 285 | 52 | 45 | 7 | - | 13 | 9 | - | 103 | 21 | 14 | 8 |
| | **G05** | - | 46 | 33 | - | - | 52 | 45 | - | - | 13 | 9 | - | 119 | 21 | 14 | 9 |
| | **M11** | - | 58 | 37 | - | 500 | 58 | 41 | 13 | - | 21 | 16 | - | 85 | 24 | 10 | 9 |
| | **M02** | - | 58 | 37 | - | 566 | 58 | 41 | 15 | - | 21 | 16 | - | 125 | 24 | 10 | 13 |
| | **M03** | - | 58 | 37 | - | 536 | 58 | 41 | 14 | - | 21 | 16 | - | 90 | 24 | 10 | 10 |
| | **M04** | - | 58 | 37 | - | 655 | 58 | 41 | 17 | - | 21 | 16 | - | 116 | 24 | 10 | 12 |
| | **M05** | - | 58 | 37 | - | 535 | 58 | 41 | 14 | - | 21 | 16 | - | 79 | 24 | 10 | 8 |
| | **M06** | - | 58 | 37 | - | 746 | 58 | 41 | 19 | - | 21 | 16 | - | 19 | 24 | 10 | 2 |
| | **H01** | - | 37 | 30 | - | 783 | 44 | 37 | 23 | 265 | 16 | 12 | 24 | 54 | 13 | 7 | 8 |

*Dessel: D1, D2 and D3 = different locations in Dessel residential areas; M1 = location at the VITO site in Mol*

*[a] Levoglucosan measurements were done in $PM_{10}$ fraction on the locations Dessel and Mol, whereas in Genk, Menen and Houtem, it was analysed in $PM_{2.5}$. Levoglucosan levels in Dessel, Mol and Houtem were 24h-averages. Those in Menen and Genk were 48h-averages*

*[b] $PM_{10}$ concentrations obtained from VMM for measuring units in: Hasselt (used as proxy for Genk, Dessel and Mol), Menen and Houtem.*

Taking all levoglucosan data of the current study and $PM_{10}$ and $PM_{2.5}$ concentrations of the available VMM measuring stations (Menen, Houtem, Hasselt), there was a significant correlation between levoglucosan and the fine particulate matter concentration (Pearson < 0.001) (

Figure 6). This was an approximation of the correlation as levoglucosan was averaged over 48h, whereas the particulate matter concentrations were 24h averages. Furthermore, PM and levoglucosan were not measured on exactly the same locations.

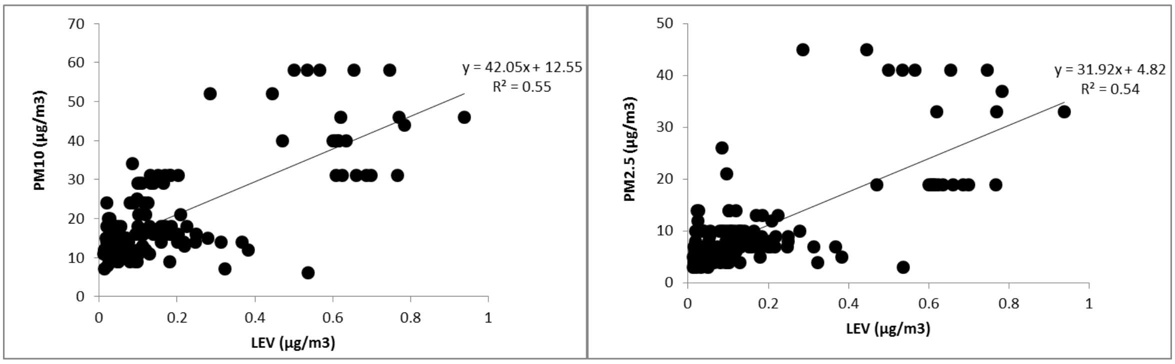

*Figure 6: Correlation between $PM_{2.5}$, $PM_{10}$ and levoglucosan measurements. All levoglucosan data of the current study (both seasons, all areas) were included. Corresponding measurements (identical period) of fine particulate matter ($PM_{10}$, $PM_{2.5}$) were obtained from nearby VMM fixed monitoring stations.*

### 3.4.4. COMPARISON WITH BAP EU GUIDELINE

A marker of PAHs in the atmosphere is the congener benzo(a)pyrene. An EU target annual average of 1 ng/m$^3$ is currently in force[6]. For the September campaign, eight 48h-measurements in Genk were higher than the annual average target value of 1 ng/m$^3$: once at the traffic station G01, six times at the traffic monitoring station G02 and once at the industry monitoring station G04. Values up to 7.0 ng/m$^3$ at G02 and 1.2 ng/m$^3$ at G04 were measured. During winter (January-February'16), G01 and G02 were not sampled anymore. At the Genk traffic monitoring station G29, a 48h-average BaP concentration of 1.3 ng/m$^3$ was measured once. However, the average BaP concentration for this monitoring station during winter was 0.3 ng/m$^3$ which was – like the 48h-average BaP concentrations of all other monitoring stations in Genk, Menen and Houtem during winter – below the target value. It should be mentioned that the current 8-weeks campaign was not representative for a yearly average concentration, and therefore not strictly comparable to the annual average target.

### 3.4.5. COMPARISON OF PAH LEVELS WITH PREVIOUS STUDIES

The levels in Genk, Menen and Houtem could be approximately compared with levels observed in other studies. An exact comparison was not possible, as the analysis techniques used, the seasons, and the (number of) PAH compounds assessed were different. This means that only the magnitude of the levels could be compared. The study of Tomaz et al. (2016) covered a year-long monitoring on a meausuring air quality station in the city of Grenoble. In that study, a total of 80 PAHs (PAHs, oxy-PAHs, nitro-PAHs) were measured in both gaseous (PUF sampling) and particulate phases ($PM_{10}$ sampling), every three days, over the whole year 2013. It is probably one of the most comprehensive studies of these compounds in ambient air. When comparing the yearly averages (of the sum of 17 common PAHs) between Grenoble and (the average of all measuring units) in the current study, the levels were comparable: respectively 22.4 vs 24.5 ng/m$^3$. The range of PAHs was overall comparable to other European locations (Table 12).

*Table 12: Comparison of total PAHs measured in the current study, with levels observed in other studies (after Tomaz et al., 2016)*

| Region | Period | Sampled phase | Type of site | # of PAHs | Mean conc (ng/m$^3$) | Reference |
|---|---|---|---|---|---|---|
| **Marseille (FR)** | summer | G+$PM_{10}$ | urban<br>suburban<br>rural | 15 | 21.4<br>4.4<br>16.0 | Albinet et al. 2007 |
| **Birmingham (UK)** | winter | G+$PM_{10}$ | traffic roadside | 17 | 87.6 | Delgado-Saborit et al. 2013 |
| **Barcelona (ES)** | all year | $PM_{10}$ | urban | 13 | 13.8 | Barrado et al. 2013 |
| **Beijing (China)** | all year | $PM_{2.5}$ | urban | 23 | 143 | Lin et al. 2015 |
| **Grenoble (FR)** | summer<br>winter<br>all year | G+$PM_{10}$ | urban | 21 | 6.3<br>41.7<br>24.5 | Tomaz et al. 2016 |
| **Flanders** | All year | G+PM | Urban+suburban+ rural | 18 | 170.4 | Wauters et al., 2008 |
| **Menen** | summer/fall + winter | G+PM | urban/suburban | 22 | 31.6$^a$ | Current study |
| **Genk** | | | urban/suburban | | 42.4$^a$ | Current study |
| **Houtem** | | | rural | | 14.2$^a$ | Current study |

*G=gas phase, $^a$: median concentrations are given, because of limited 4-weaks sampling in each season, on several different measuring locations in each of the regions*

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[6] http://ec.europa.eu/environment/air/quality/standards.htm

### 3.4.6. PAH PROFILES INDICATING RELATIVE CONTRIBUTION INDIVIDUAL PAH TO TOTAL PAH

Overall the lower molecular weight PAHs (NAP, 1Me- and 2MeNAP) made up the largest proportion of all measured PAHs on all sampling locations, in winter as well as in summer (between 78.5% and 93.9%) (Figure 7). There was a (slight) seasonal shift to lower molecular weight PAHs being relatively more present in the PAH profile of the winter compared to summer-fall. This shift was most clear in Genk, where the proportion of low molecular weight PAHs increased from 78.5% to 92.2% of all PAHs (Figure 7).

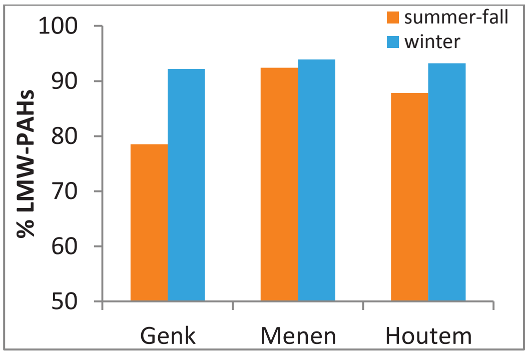

*Figure 7: seasonal variation in proportion of low molecular weight PAHs (LMW-PAHs: NAP+2MeNAP+1MeNAP+ACY+ACE+FLU+PHE+9MePHE+ANT+2MeANT) to the mass of all 22 PAHs measured. The average contribution of the 4 weeks campaign and all measuring locations in respectively Genk, Menen and Houtem are shown*

On the measuring station G04, in summer-fall the relative contribution of ACE (3.9% of total PAHs) and ACY (even up to 28.7%) was much higher than on other measuring units. In winter this was not anymore seen (respectively 0.9 and 2.5% of total PAHs for ACE and ACY) (Table 14, Table 15).

The contribution of the different PAHs could be compared to the earlier mentioned publication on PAH measurements in the ambient air particle and gas phase from Grenoble (France) (Tomaz et al., 2016). Not all 21 parent PAH compounds measured by Tomaz et al. (2016) were similar to the 22 PAHs measured in the current study[7]. In Grenoble the yearly annual concentrations of PHE, 2MeNAP, PYR and FLA accounted for 74% of the total PAHs. This was also seen in our study in case NAP, ACY, 9MePHE, 2MeANT and PIC as in Grenoble study, were excluded from the sum of the PAHs: 51% Genk, 57% Menen, 57% Houtem. The most important difference between both studies, was 1-MeNAP, which was very apparent in our study, but marginally present in the Grenoble campaign.

The correlations among levoglucosan and/or different subsets of PAHs were all significant, except for the correlation between levoglucosan and the sum of the 22 PAHs. Levoglucosan was, as expected, best correlated with low molecular PAHs (r=0.19, p=0.018), and the sum of NAP, PHE, FLA and PYR (r=0.21, p=0.009), that are typically formed at lower temperatures during biomass burning (Khalili et al., 1995; Taxeira et al., 2013).

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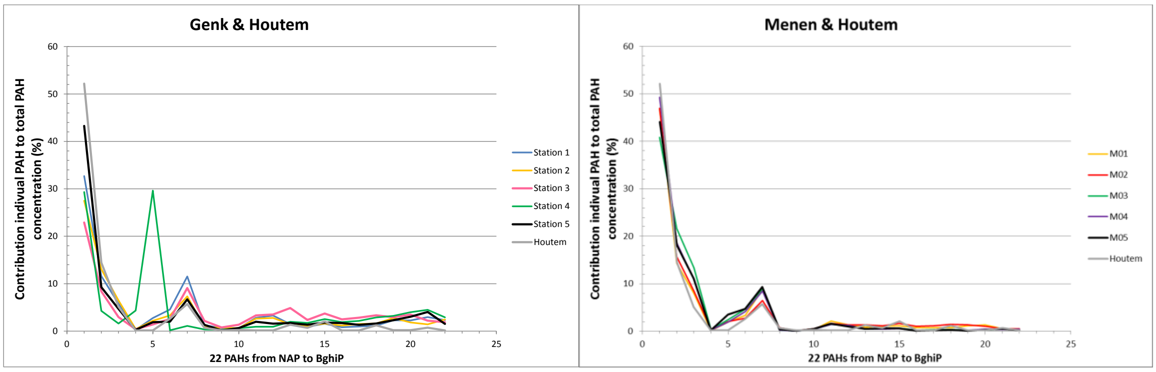
[7] PAHs that were measured in the study of Tomaz et al. (2016), but not in our study are 2-methyl fuoranthene (2MeFLA), Retene (RET) and benzo[j]fuoranthene (BjF). PAHs measured in our study and not in the study of Tomaz et al. (2016), were NAP, ACY, 9MePHE, 2MeANT and PIC.

*Table 13: Spearman rank correlations between different subsets of PAHs analyzed in the current study over all measuring locations and both season*

| Compounds | N | Spearman rank correlation | p-value |
|---|---|---|---|
| LEV & LMW-PAH | 146 | 0.23 | 0.0049 |
| LEV& biomass-PAH | 146 | 0.29 | 0.0004 |
| LEV & HMW-PAH | 146 | -0.17 | 0.0446 |
| LEV & sum_PAH | 146 | 0.20 | 0.0147 |
| LMW-PAH & sum_PAH | 168 | 0.98 | 0.0000 |
| HMW-PAH & sum_PAH | 168 | 0.59 | 0.0000 |
| LMW-PAH & HMW-PAH | 168 | 0.48 | 0.0000 |

*with: biomass PAH= NAP+PHE+FLA+PYR; LMW-PAH= low molecular weight PAHs =* NAP+2MeNAP+ 1MeNAP+ACY+ACE+FLU+PHE+9MePHE+ANT+2MeANT *HMW-PAH=high molecular weight PAHs = BaA+CHR+BbF+ BkF+BeP+BaP+IcdP+DahA+PIC+BghiP, sum_PAH=sum of all 22 PAHs measured in the current study.*

**Summer-fall**



**Winter**

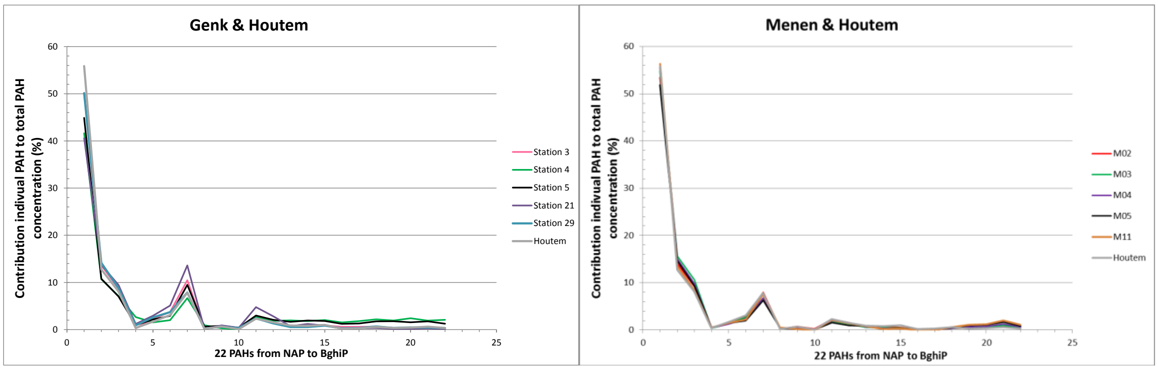


*Figure 8: Profile of PAHs at all measuring stations in Genk, Menen and Houtem, in summer-fall (upper row), and winter (lower row). The X-axis gives the measured PAHs with increasing number of benzene rings from left to right. 1=NAP; 2=2MeNAP; 3=1MeNAP; 4=ACY; 5=ACE; 6=FLU; 7=PHE; 8=9MePHE; 9=ANT; 10=2MeANT; 11=FLA; 12=PYR; 13=BaA; 14=CHR; 15=BbF; 16=BkF; 17=BeP; 18=BaP; 19=IcdP; 20=DahA; 21=PIC; 22=BghiP. Y-axis: concentration of the different PAHs as % of total sum of PAHs*

*Table 14 Contribution of individual PAH to total PAH concentration (in %, based on average values per monitoring station) for summer-fall period (Sept’15)*

| Location | Station | PAH(%) NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Genk | G01 | 32.5 | 14.6 | 7.5 | 1.1 | 3.7 | 4.7 | 10.7 | 1.1 | 0.6 | 0.6 | 2.6 | 3.3 | 1.2 | 0.7 | 1.2 | 0.7 | 1.0 | 1.0 | 2.6 | 2.7 | 3.5 | 2.4 |
|  | G02 | 29.7 | 11.9 | 6.5 | 0.6 | 3.7 | 4.2 | 8.0 | 0.9 | 0.6 | 0.7 | 3.3 | 3.7 | 1.6 | 1.5 | 2.1 | 1.7 | 2.1 | 2.2 | 3.3 | 3.6 | 4.7 | 3.4 |
|  | G03 | 42.9 | 12.4 | 6.4 | 0.4 | 2.5 | 3.6 | 9.8 | 1.4 | 0.5 | 0.7 | 2.6 | 2.4 | 2.1 | 1.2 | 1.9 | 1.1 | 1.2 | 1.5 | 1.7 | 1.7 | 1.1 | 1.2 |
|  | G04 | 30.2 | 4.4 | 1.7 | 3.9 | 28.7 | 0.2 | 1.2 | 0.5 | 0.4 | 0.6 | 1.0 | 1.0 | 2.0 | 1.7 | 2.4 | 1.9 | 2.1 | 2.7 | 3.3 | 3.9 | 3.8 | 2.6 |
|  | G05 | 43.5 | 11.0 | 6.3 | 0.7 | 2.3 | 3.4 | 9.6 | 0.9 | 0.2 | 0.6 | 2.0 | 1.6 | 1.7 | 1.2 | 1.9 | 1.3 | 1.5 | 1.8 | 2.0 | 2.5 | 2.8 | 1.2 |
| Menen | M01 | 44.8 | 16.3 | 9.3 | 0.3 | 2.5 | 4.1 | 9.1 | 0.5 | 0.1 | 0.4 | 2.1 | 1.3 | 0.9 | 1.0 | 1.2 | 0.7 | 0.8 | 0.9 | 1.0 | 1.2 | 1.0 | 0.6 |
|  | M02 | 43.1 | 17.7 | 9.9 | 0.4 | 2.2 | 3.8 | 7.5 | 0.7 | 0.1 | 0.5 | 1.7 | 1.4 | 1.3 | 1.1 | 1.5 | 0.9 | 1.1 | 1.2 | 1.2 | 1.1 | 0.9 | 0.7 |
|  | M03 | 43.3 | 19.8 | 13.1 | 0.5 | 2.3 | 4.4 | 8.8 | 0.4 | 0.2 | 0.5 | 1.5 | 1.4 | 0.6 | 0.5 | 0.6 | 0.2 | 0.3 | 0.3 | 0.3 | 0.2 | 0.4 | 0.3 |
|  | M04 | 50.0 | 17.7 | 11.2 | 0.3 | 2.0 | 4.0 | 8.1 | 0.5 | 0.0 | 0.5 | 1.5 | 1.3 | 0.5 | 0.4 | 0.6 | 0.1 | 0.2 | 0.3 | 0.2 | 0.3 | 0.3 | 0.2 |
|  | M05 | 43.6 | 17.7 | 11.6 | 0.5 | 4.1 | 4.9 | 9.8 | 0.5 | 0.4 | 0.5 | 1.8 | 1.3 | 0.5 | 0.5 | 0.7 | 0.1 | 0.4 | 0.4 | 0.1 | 0.2 | 0.4 | 0.1 |
| Houtem |  | 55.4 | 14.5 | 6.5 | 0.1 | 0.2 | 2.1 | 6.1 | 1.1 | 0.2 | 0.3 | 0.4 | 0.5 | 1.3 | 0.8 | 2.3 | 0.3 | 0.2 | 1.2 | 2.7 | 0.4 | 0.6 | 2.7 |

*Table 15 Contribution of individual PAH to total PAH concentration (in %, based on average values per monitoring station) for winter period (Jan-Feb’16)*

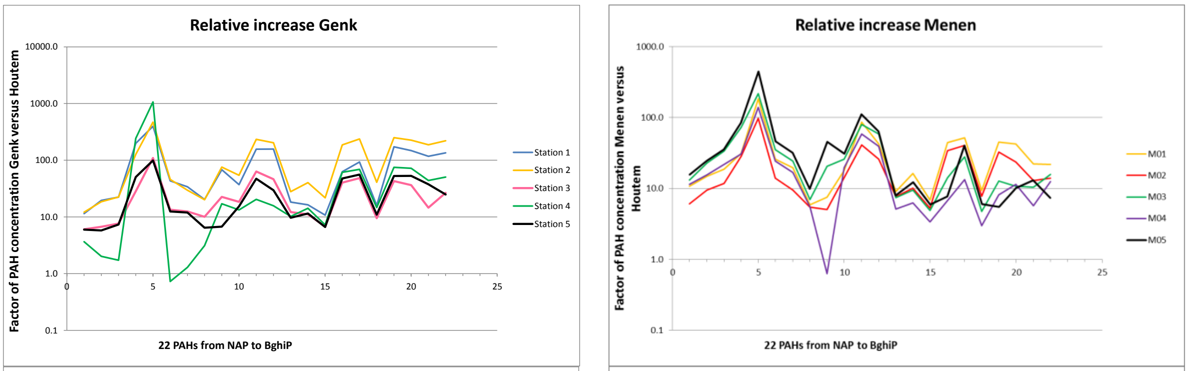
| Location | Station | PAH(%) NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Genk | G21 | 37.7 | 14.3 | 10.1 | 1.6 | 2.9 | 5.1 | 13.7 | 0.1 | 1.2 | 0.4 | 5.2 | 3.2 | 0.9 | 1.4 | 0.9 | 0.3 | 0.3 | 0.4 | 0.1 | 0.1 | 0.2 | 0.1 |
|  | G29 | 44.1 | 14.5 | 9.5 | 2.3 | 2.5 | 4.4 | 11.4 | 0.1 | 0.8 | 0.4 | 4.2 | 2.4 | 0.2 | 0.2 | 0.8 | 0.3 | 0.4 | 0.6 | 0.3 | 0.2 | 0.3 | 0.3 |
|  | G03 | 51.9 | 13.1 | 8.5 | 0.3 | 2.1 | 3.6 | 9.6 | 0.2 | 0.7 | 0.3 | 2.7 | 1.8 | 0.6 | 0.8 | 0.9 | 0.5 | 0.6 | 0.6 | 0.4 | 0.3 | 0.3 | 0.3 |
|  | G04 | 53.9 | 11.2 | 7.5 | 2.5 | 0.9 | 1.7 | 6.0 | 0.4 | 0.3 | 0.2 | 2.2 | 1.5 | 1.1 | 1.2 | 1.2 | 0.8 | 1.0 | 1.3 | 1.4 | 1.2 | 1.3 | 1.2 |
|  | G05 | 46.9 | 12.2 | 7.8 | 0.5 | 2.1 | 3.2 | 10.3 | 0.3 | 0.7 | 0.4 | 3.2 | 2.0 | 1.0 | 1.5 | 1.3 | 0.7 | 0.9 | 1.1 | 1.0 | 0.9 | 1.0 | 0.8 |
| Menen | M11 | 56.5 | 13.5 | 8.6 | 0.5 | 1.5 | 2.3 | 7.6 | 0.4 | 0.4 | 0.1 | 2.1 | 1.4 | 0.7 | 0.4 | 0.3 | 0.0 | 0.0 | 0.4 | 0.5 | 0.9 | 1.4 | 0.5 |
|  | M02 | 53.0 | 14.9 | 9.9 | 1.4 | 1.4 | 2.8 | 8.0 | 0.2 | 0.7 | 0.2 | 2.2 | 1.7 | 0.6 | 0.7 | 0.5 | 0.1 | 0.1 | 0.3 | 0.2 | 0.3 | 0.5 | 0.2 |
|  | M03 | 55.4 | 15.2 | 10.8 | 0.4 | 1.3 | 2.7 | 6.7 | 0.2 | 0.5 | 0.1 | 1.8 | 1.2 | 0.6 | 0.6 | 0.5 | 0.1 | 0.0 | 0.3 | 0.3 | 0.4 | 0.7 | 0.3 |
|  | M04 | 55.4 | 15.1 | 9.8 | 0.6 | 1.2 | 2.4 | 7.1 | 0.2 | 0.5 | 0.1 | 2.0 | 1.5 | 0.6 | 0.5 | 0.4 | 0.0 | 0.0 | 0.3 | 0.3 | 0.5 | 0.9 | 0.3 |
|  | M05 | 53.0 | 15.5 | 9.8 | 0.5 | 1.7 | 2.2 | 6.9 | 0.3 | 0.3 | 0.1 | 2.3 | 1.5 | 1.1 | 0.9 | 0.4 | 0.0 | 0.0 | 0.4 | 0.5 | 0.8 | 1.3 | 0.5 |
| Houtem |  | 55.7 | 12.4 | 8.3 | 0.3 | 1.6 | 3.2 | 8.1 | 0.2 | 0.6 | 0.1 | 2.5 | 1.7 | 0.7 | 0.8 | 1.0 | 0.2 | 0.3 | 0.6 | 0.3 | 0.5 | 0.7 | 0.3 |

### 3.4.7. PAH LEVELS IN GENK AND MENEN COMPARED TO REFERENCE STATION HOUTEM

In Menen, Genk and Houtem, PAHs were measured at the same days, during 4 weeks in summe-fall and 4 weeks in winter. The concentrations on the locations of Menen and Genk could therefore be directly related to the levels in rural area Houtem, which was a background station in the West of Flanders (about 20 km from the Nordsea coast). Genk and Menen were more urbanized semi-urban areas of Flanders.

Overall, in the **summer-fall** period, the levels of the individual compounds measured on the different stations of Genk and Menen were mostly higher than those in Houtem. However, industry station G04 and traffic station M05 had some lower molecular weight PAHs, with similar concentrations as for Houtem. In **winter** most of the stations in Genk and Menen had comparable or lower PAH levels in comparison to Houtem. Only the traffic station G21, G29, and the industry station G03 showed higher concentrations for most of the PAHs than the levels in Houtem measured in that same period.

**Summer-fall**



**Winter**

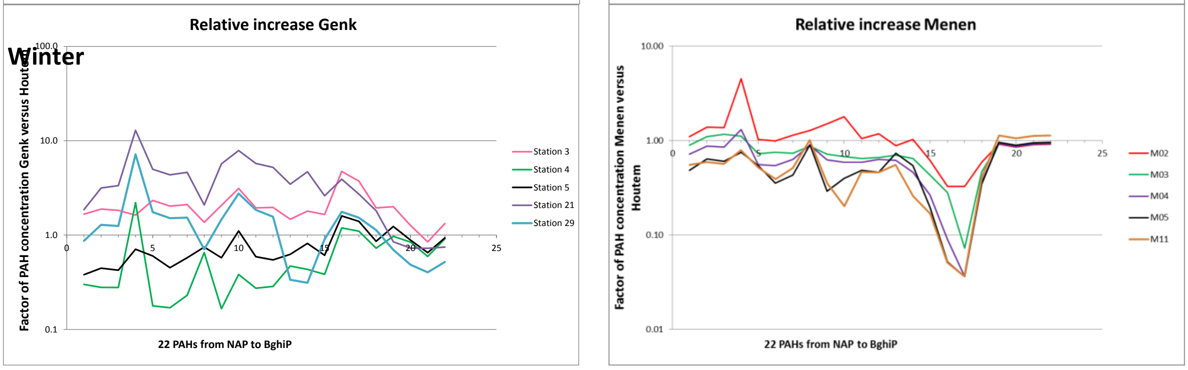


*Figure 9: PAH profiles for the monitoring stations of Genk (left) and Menen (right) relatively to the monitoring station of Houtem, pictured for summer-fall (upper row) and winter (lower row): The X-axis gives the measured PAHs with increasing number of benzene rings from left to right. 1=NAP; 2=2MeNAP; 3=1MeNAP; 4=ACY; 5=ACE; 6=FLU; 7=PHE; 8=9MePHE; 9=ANT; 10=2MeANT; 11=FLA; 12=PYR; 13=BaA; 14=CHR; 15=BbF; 16=BkF; 17=BeP; 18=BaP; 19=IcdP; 20=DahA; 21=PIC; 22=BghiP. The Y-axis gives the ratio of the average concentration in the individual measuring stations of Genk/Diepenbeek or Menen compared to Houtem.*

### 3.4.8. PAH AND LEV LEVELS ON THE INDIVIDUAL SAMPLING STATIONS

The individual PAH and levoglucosan levels on each of the different sampling time points and over the different seasons were be compared to evaluate possible local and/or periodic sources or influences of meteorological conditions. In Table 16,Table 17, Table 18 and Table 19, Table 20 and Table 21 the PAH and levoglucosan concentrations measured during respectively the summer-fall and the winter campaign, are given for Genk/Diepenbeek, Menen and Houtem. The PAHs are arranged from left to right according to increasing molecular weight as listed in Table 2.

| ID | start | NA | 2Me | 1Me | AC | AC | FL | PH | 9Me | AN | 2Me | FL | PY | Ba | CH | Bb | Bk | Be | Ba | Icd | Da | PIC | Bg | LE |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| M11 indus | 21/1/16 2:23 PM | 29.662 | 7.564 | 4.451 | 0.169 | 0.692 | 1.327 | 3.977 | 0.123 | 0.229 | 0.060 | 1.074 | 0.725 | 0.251 | 0.219 | 0.118 | 0.005 | 0.005 | 0.117 | 0.125 | 0.247 | 0.401 | 0.125 | 500 |
| | 26/1 | 9.1 | 2.11 | 1.35 | 0.0 | 0.3 | 0.4 | 1.6 | 0.08 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 8 |
| | 28/1 | 9.7 | 1.49 | 1.38 | 0.0 | 0.2 | 0.3 | 1.3 | 0.08 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 9 |
| | 2/2/ | 7.5 | 1.84 | 1.12 | 0.0 | 0.2 | 0.1 | 0.7 | 0.08 | 0.0 | 0.00 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 2 |
| | 4/2/ | 8.7 | 2.24 | 1.37 | 0.0 | 0.3 | 0.3 | 1.4 | 0.08 | 0.0 | 0.00 | 0.3 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 1 |
| | 9/2/ | 12. | 2.91 | 1.87 | 0.1 | 0.3 | 0.4 | 1.4 | 0.10 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 3 |
| | 11/2 | 23. | 5.68 | 3.66 | 0.3 | 0.6 | 1.0 | 3.1 | 0.12 | 0.1 | 0.02 | 0.9 | 0.7 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| | 16/2 | 22. | 5.54 | 3.58 | 0.1 | 0.5 | 0.9 | 2.8 | 0.10 | 0.2 | 0.00 | 0.8 | 0.6 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| M02 indus | 21/1 | 54. | 16.0 | 10.2 | 0.3 | 1.2 | 3.0 | 8.7 | 0.17 | 0.8 | 0.32 | 2.4 | 1.8 | 0.4 | 0.7 | 0.4 | 0.0 | 0.0 | 0.2 | 0.1 | 0.1 | 0.3 | 0.1 | 5 |
| | 26/1 | 18. | 3.98 | 3.08 | 0.1 | 0.5 | 0.9 | 2.8 | 0.08 | 0.1 | 0.05 | 0.8 | 0.5 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 28/1 | 18. | 4.73 | 3.18 | 3.1 | 0.4 | 0.8 | 2.6 | 0.06 | 0.2 | 0.08 | 0.7 | 0.4 | 0.2 | 0.2 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 2/2/ | 20. | 4.69 | 2.99 | 0.1 | 0.5 | 0.8 | 2.5 | 0.08 | 0.1 | 0.02 | 0.7 | 0.5 | 0.2 | 0.2 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 2 |
| | 4/2/ | 22. | 5.94 | 3.76 | 0.1 | 0.5 | 1.2 | 3.6 | 0.09 | 0.2 | 0.08 | 0.9 | 0.6 | 0.1 | 0.2 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 2 |
| | 9/2/ | 25. | 6.04 | 3.98 | 0.1 | 0.6 | 1.0 | 3.2 | 0.17 | 0.3 | 0.08 | 0.8 | 0.6 | 0.2 | 0.3 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 3 |
| | 11/2 | 40. | 12.7 | 8.57 | 1.5 | 1.1 | 2.1 | 6.0 | 0.16 | 0.5 | 0.15 | 1.5 | 1.2 | 0.4 | 0.6 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
| | 16/2 | 44. | 14.7 | 9.80 | 0.5 | 1.4 | 2.5 | 7.3 | 0.15 | 0.8 | 0.22 | 2.2 | 1.8 | 0.6 | 0.8 | 0.5 | 0.1 | 0.0 | 0.2 | 0.1 | 0.1 | 0.2 | 0.1 | 7 |
| M03 back | 21/1 | 43. | 12.0 | 8.04 | 0.2 | 0.8 | 2.1 | 5.9 | 0.12 | 0.3 | 0.19 | 1.4 | 1.0 | 0.3 | 0.4 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 5 |
| | 26/1 | 12. | 3.03 | 2.43 | 0.0 | 0.4 | 0.8 | 2.0 | 0.07 | 0.2 | 0.00 | 0.5 | 0.3 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 9 |
| | 28/1 | 16. | 1.84 | 2.95 | 0.1 | 0.5 | 0.8 | 2.3 | 0.08 | 0.1 | 0.02 | 0.5 | 0.3 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 2/2/ | 15. | 5.17 | 3.71 | 0.1 | 0.4 | 0.6 | 1.6 | 0.08 | 0.1 | 0.00 | 0.5 | 0.3 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 9 |
| | 4/2/ | 16. | 4.85 | 3.17 | 0.0 | 0.3 | 0.8 | 2.2 | 0.07 | 0.1 | 0.00 | 0.6 | 0.3 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 9/2/ | 19. | 5.96 | 4.13 | 0.1 | 0.4 | 0.9 | 2.0 | 0.07 | 0.1 | 0.00 | 0.5 | 0.3 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 11/2 | 35. | 9.59 | 6.44 | 0.5 | 0.7 | 1.4 | 3.5 | 0.09 | 0.2 | 0.07 | 0.9 | 0.6 | 0.3 | 0.4 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| | 16/2 | 38. | 11.7 | 7.79 | 0.1 | 0.6 | 1.7 | 4.0 | 0.09 | 0.3 | 0.07 | 1.2 | 0.9 | 0.5 | 0.6 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| M04 back | 21/1 | 38. | 9.68 | 6.54 | 0.1 | 0.7 | 1.9 | 5.6 | 0.11 | 0.3 | 0.15 | 1.6 | 1.1 | 0.3 | 0.4 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
| | 26/1 | 10. | 3.17 | 1.99 | 0.0 | 0.2 | 0.4 | 1.5 | 0.07 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 28/1 | 12. | 3.12 | 1.90 | 0.0 | 0.2 | 0.4 | 1.3 | 0.06 | 0.0 | 0.00 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 2/2/ | 11. | 3.36 | 2.11 | 0.0 | 0.3 | 0.3 | 1.1 | 0.07 | 0.0 | 0.00 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 4/2/ | 12. | 3.48 | 2.11 | 0.1 | 0.2 | 0.5 | 1.8 | 0.07 | 0.0 | 0.00 | 0.5 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 9/2/ | 15. | 4.13 | 2.67 | 0.1 | 0.3 | 0.5 | 1.6 | 0.07 | 0.0 | 0.00 | 0.4 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 2 |
| | 11/2 | 27. | 7.45 | 4.96 | 0.7 | 0.6 | 1.1 | 3.2 | 0.10 | 0.2 | 0.06 | 0.9 | 0.7 | 0.3 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
| | 16/2 | 29. | 8.93 | 5.92 | 0.4 | 0.6 | 1.4 | 3.8 | 0.12 | 0.3 | 0.09 | 1.1 | 0.8 | 0.3 | 0.4 | 0.1 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
| M05 traffi | 21/1 | 29. | 7.97 | 5.15 | 0.2 | 0.6 | 1.2 | 3.9 | 0.09 | 0.2 | 0.11 | 1.0 | 0.7 | 0.2 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 5 |
| | 26/1 | 9.0 | 4.76 | 2.99 | 0.1 | 0.4 | 0.4 | 1.3 | 0.07 | 0.0 | 0.00 | 0.3 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 7 |
| | 28/1 | 1.4 | 0.36 | 0.20 | 0.0 | 0.0 | 0.0 | 0.0 | 0.07 | 0.0 | 0.00 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 2/2/ | 11. | 2.75 | 1.70 | 0.0 | 0.3 | 0.3 | 1.2 | 0.07 | 0.0 | 0.00 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 1 |
| | 4/2/ | 7.4 | 2.30 | 1.48 | 0.1 | 0.1 | 0.2 | 0.9 | 0.07 | 0.0 | 0.00 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 9/2/ | 16. | 3.82 | 2.44 | 0.1 | 0.3 | 0.4 | 1.6 | 0.09 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 2 |
| | 11/2 | 4.9 | 1.52 | 0.94 | 0.1 | 0.1 | 0.0 | 0.4 | 0.10 | 0.0 | 0.00 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 4 |
| | 16/2 | 27. | 8.03 | 5.06 | 0.1 | 1.2 | 1.6 | 4.4 | 0.11 | 0.3 | 0.08 | 2.3 | 1.4 | 1.0 | 1.3 | 0.2 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| M06 traffi | 21/1 | 7.3 | 1.62 | 1.06 | 0.8 | 0.1 | 0.1 | 0.7 | 0.09 | 0.1 | 0.00 | 0.2 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 7 |
| | 26/1 | Technical problem PAH measurment | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 28/1 | | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 2/2/ | | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 4/2/ | | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 9/2/ | | | | | | | | | | | | | | | | | | | | | | | 2 |
| | 11/2 | | | | | | | | | | | | | | | | | | | | | | | 5 |
| | 16/2 | | | | | | | | | | | | | | | | | | | | | | | 6 |

Increases in levoglucosan were consistently observed in the colder periods (19/1, 21/1, 11/2 and 16/2/16) reaching levels in the range of approximately 300 to 900 ng/m$^3$. Equal high values were attained on the different measuring locations in Menen. In Genk the industry station G03, and the background station G05 showed somewhat lower levoglucosan concentrations. The PAHs showed a more locally diverse pattern:

*Genk/Diepenbeek*

The **traffic monitoring stations G01** and **G02** showed rather consistently relatively higher levels of low molecular and high molecular weight PAHs. Both stations were located in major heavy traffic roads: respectively 17.10$^3$ and >20.10$^3$ vehicles/day, of with around respectively 2000 and 1000 heavy traffic vehicles/day. These stations were not anymore sampled in the winter time, and needed to be replaced by two nearby sampling stations **G21** and **G29**. G21 was also located in a major road with heavy traffic (>20.10$^3$ vehicles/day of which around 2000 heavy vehicles/day). Whereas the G29 location was a major road with about 11000 veh/day, but without heavy traffic. G21 showed rather high and G29 somewhat variable low- to medium-molecular weight PAHs (NAP to PYR). On both locations much lower concentrations of the 5-rings IcdP, DahA, PIC and the 6-ring BghiP were observed, compared to the summer stations G01 and G02.

At **G03**, a station W of the industrial area Genk-South, relatively high values of medium to high molecular weight PAHs (4- to 6-rings) were measured in the summer fall campaign, as was also the case for G04 located N of the industrial area Genk-South. On some summer-fall days (15/9 and 22/9), the 2- and 3-ring PAHs from NAP to PHE were also relatively high at G03 (Table 14). During the winter campaign the 4-ring and some 5-ring PAHs were still relatively high at the measuring location G03, and the low molecular PAHs were now clearly present at all winter sampling days (Table 19).
As mentioned above, on the industry monitoring station **G04**, relative high values of medium to high molecular weight PAHs (4- to 6-ring PAHs) were measured in the summer-fall campaign. In the winter time that was not anymore observed. In summer-fall also high concentrations of the 2-ring compounds ACY, ACE were the highest measured in the campaigns, whereas all other low molecular PAHs were low (Table 16). In winter there were two days, on which the temperature dropped to zero (19/1 and 21/1). Only on those days all lower PAHs appeared clearly in the PAH spectrum. ACY and certainly ACE did not anymore appear to the extend as observed in summer.

The background station **G05** showed similar concentrations of medium to high molecular weight PAHs observed in the industry monitoring stations G03 and even more similar to G04. G05 also showed tendency of increased ACE and ACY in summer-fall, which was somewhat reduced to slightly increased tendency of ACY in winter. Measuring units G04 and G05 were located 2 km away from each other respectively at N and N-E of Genk-South industry. Both G04 and G05 were located in streets without heavy traffic, with respectively around 4000 and 1500 vehicles/day.

*Menen*

The industry stations **M01** and **M02** (W and N-W of Menen industrial area) showed a similar pattern of relatively high levels of 4- and 5-rings (Table 17), and higher appearance of low molecular PAHs (NAP, 2MeNAP, 1MeNAP, ACE, PHE) in the first sampling days of the summer-fall campaign (1/9-10/9). In the next sampling period (15/9-24/9), those PAHs were low, and only 9MePHE appeared relatively strong in the PAH spectrum. The **M11** station, (which replaced the M01 station of the summer-fall period) mainly showed the 5- and 6 rings IcdP, DahA, PIC and BghiP, and some lower molecular weight PAHs, in the period 9-16/2, when levoglucosan was increased. On the location **M02,** also the lower molecular weight concentrations were higher than in the summer period, and also here as for M11, especially in the period of 9-16/2, when levoglucosan was increased (Table 20).

The background stations **M03** and **M04** located at the East side of the industry terrains of Menen, showed in summer-fall relatively high levels of the low molecular PAHs (NAP, 2MeNAP, 1MeNAP, ACE, PHE). It was very striking that in the same period from 15-24/9, also here 9MePHE was clearly present, as was observed for M01 and M02 in that sampling period (Table 17). In the winter period M03 and M04 mainly showed lower molecular weight PAHs, with the highest concentrations from 9-16/2.

On the traffic location **M05** with around 9000 vehicles per day, of which about 600 heavy traffic vehicles per day, all 2-, 3- and 4-ring PAHs were clearly measurable. The traffic station **M06** had only limited valuable measurements, but it could be seen that the pattern was different, with low concentrations of all low molecular weight PAHs, except 9MePHE, whereas the 4-ring BaA, BbF, BkF, and the 5-ring BeP and BaP were relatively present as on the industry M01 and M02 and the background station M03 (Table 17). In the winter time, there were lower PAH levels measured on the traffic locations, except for some days at location M05, e.g. 16/2, where clearly higher levels of 2-, 3- and 4-rings were observed, together with high levoglucosan concentrations (608.2 ng/m$^3$) (Table 20).

*Houtem*

In summer, all levels of PAHs were low compared to the locations Menen and Genk. Only in the second half of September, the levels of 9MePHE were relatively higher, possibly associated with the higher levoglucosan concentration measured on 25/9. Also in the wintertime higher PAH levels were visible in the PAH pattern, mainly when relatively high levels of levoglucosan were measured (

| ID | start | NA | 2Me | 1Me | AC | AC | FL | PH | 9Me | AN | 2Me | FL | PY | Ba | CH | Bb | Bk | Be | Ba | Icd | Da | PIC | Bg | LE |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| M11 | 21/1/16 2:23 PM | 29.662 | 7.564 | 4.451 | 0.169 | 0.692 | 1.327 | 3.977 | 0.123 | 0.229 | 0.060 | 1.074 | 0.725 | 0.251 | 0.219 | 0.118 | 0.005 | 0.005 | 0.117 | 0.125 | 0.247 | 0.401 | 0.125 | 500 |
| indus | 26/1 | 9.1 | 2.11 | 1.35 | 0.0 | 0.3 | 0.4 | 1.6 | 0.08 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 8 |
|  | 28/1 | 9.7 | 1.49 | 1.38 | 0.0 | 0.2 | 0.3 | 1.3 | 0.08 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 9 |
|  | 2/2/ | 7.5 | 1.84 | 1.12 | 0.0 | 0.2 | 0.1 | 0.7 | 0.08 | 0.0 | 0.00 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 2 |
|  | 4/2/ | 8.7 | 2.24 | 1.37 | 0.0 | 0.3 | 0.3 | 1.4 | 0.08 | 0.0 | 0.00 | 0.3 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 1 |
|  | 9/2/ | 12. | 2.91 | 1.87 | 0.1 | 0.3 | 0.4 | 1.4 | 0.10 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 3 |
|  | 11/2 | 23. | 5.68 | 3.66 | 0.3 | 0.6 | 1.0 | 3.1 | 0.12 | 0.1 | 0.02 | 0.9 | 0.7 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
|  | 16/2 | 22. | 5.54 | 3.58 | 0.1 | 0.5 | 0.9 | 2.8 | 0.10 | 0.2 | 0.00 | 0.8 | 0.6 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| M02 | 21/1 | 54. | 16.0 | 10.2 | 0.3 | 1.2 | 3.0 | 8.7 | 0.17 | 0.8 | 0.32 | 2.4 | 1.8 | 0.4 | 0.7 | 0.4 | 0.0 | 0.0 | 0.2 | 0.1 | 0.1 | 0.3 | 0.1 | 5 |
| indus | 26/1 | 18. | 3.98 | 3.08 | 0.1 | 0.5 | 0.9 | 2.8 | 0.08 | 0.1 | 0.05 | 0.8 | 0.5 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 28/1 | 18. | 4.73 | 3.18 | 3.1 | 0.4 | 0.8 | 2.6 | 0.06 | 0.2 | 0.08 | 0.7 | 0.4 | 0.2 | 0.2 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 2/2/ | 20. | 4.69 | 2.99 | 0.1 | 0.5 | 0.8 | 2.5 | 0.08 | 0.1 | 0.02 | 0.7 | 0.5 | 0.2 | 0.2 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 2 |
|  | 4/2/ | 22. | 5.94 | 3.76 | 0.1 | 0.5 | 1.2 | 3.6 | 0.09 | 0.2 | 0.08 | 0.9 | 0.6 | 0.1 | 0.2 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 2 |
|  | 9/2/ | 25. | 6.04 | 3.98 | 0.1 | 0.6 | 1.0 | 3.2 | 0.17 | 0.3 | 0.08 | 0.8 | 0.6 | 0.2 | 0.3 | 0.2 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 3 |
|  | 11/2 | 40. | 12.7 | 8.57 | 1.5 | 1.1 | 2.1 | 6.0 | 0.16 | 0.5 | 0.15 | 1.5 | 1.2 | 0.4 | 0.6 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
|  | 16/2 | 44. | 14.7 | 9.80 | 0.5 | 1.4 | 2.5 | 7.3 | 0.15 | 0.8 | 0.22 | 2.2 | 1.8 | 0.6 | 0.8 | 0.5 | 0.1 | 0.0 | 0.2 | 0.1 | 0.1 | 0.2 | 0.1 | 7 |
| M03 | 21/1 | 43. | 12.0 | 8.04 | 0.2 | 0.8 | 2.1 | 5.9 | 0.12 | 0.3 | 0.19 | 1.4 | 1.0 | 0.3 | 0.4 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 5 |
| back | 26/1 | 12. | 3.03 | 2.43 | 0.0 | 0.4 | 0.8 | 2.0 | 0.07 | 0.2 | 0.00 | 0.5 | 0.3 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 9 |
|  | 28/1 | 16. | 1.84 | 2.95 | 0.1 | 0.5 | 0.8 | 2.3 | 0.08 | 0.1 | 0.02 | 0.5 | 0.3 | 0.1 | 0.0 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 2/2/ | 15. | 5.17 | 3.71 | 0.1 | 0.4 | 0.6 | 1.6 | 0.08 | 0.1 | 0.00 | 0.5 | 0.3 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 9 |
|  | 4/2/ | 16. | 4.85 | 3.17 | 0.0 | 0.3 | 0.8 | 2.2 | 0.07 | 0.1 | 0.00 | 0.6 | 0.3 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 9/2/ | 19. | 5.96 | 4.13 | 0.1 | 0.4 | 0.9 | 2.0 | 0.07 | 0.1 | 0.00 | 0.5 | 0.3 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 11/2 | 35. | 9.59 | 6.44 | 0.5 | 0.7 | 1.4 | 3.5 | 0.09 | 0.2 | 0.07 | 0.9 | 0.6 | 0.3 | 0.4 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
|  | 16/2 | 38. | 11.7 | 7.79 | 0.1 | 0.6 | 1.7 | 4.0 | 0.09 | 0.3 | 0.07 | 1.2 | 0.9 | 0.5 | 0.6 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| M04 | 21/1 | 38. | 9.68 | 6.54 | 0.1 | 0.7 | 1.9 | 5.6 | 0.11 | 0.3 | 0.15 | 1.6 | 1.1 | 0.3 | 0.4 | 0.3 | 0.0 | 0.0 | 0.1 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
| back | 26/1 | 10. | 3.17 | 1.99 | 0.0 | 0.2 | 0.4 | 1.5 | 0.07 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 28/1 | 12. | 3.12 | 1.90 | 0.0 | 0.2 | 0.4 | 1.3 | 0.06 | 0.0 | 0.00 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 2/2/ | 11. | 3.36 | 2.11 | 0.0 | 0.3 | 0.3 | 1.1 | 0.07 | 0.0 | 0.00 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 4/2/ | 12. | 3.48 | 2.11 | 0.1 | 0.2 | 0.5 | 1.8 | 0.07 | 0.0 | 0.00 | 0.5 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 9/2/ | 15. | 4.13 | 2.67 | 0.1 | 0.3 | 0.5 | 1.6 | 0.07 | 0.0 | 0.00 | 0.4 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 2 |
|  | 11/2 | 27. | 7.45 | 4.96 | 0.7 | 0.6 | 1.1 | 3.2 | 0.10 | 0.2 | 0.06 | 0.9 | 0.7 | 0.3 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
|  | 16/2 | 29. | 8.93 | 5.92 | 0.4 | 0.6 | 1.4 | 3.8 | 0.12 | 0.3 | 0.09 | 1.1 | 0.8 | 0.3 | 0.4 | 0.1 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 6 |
| M05 | 21/1 | 29. | 7.97 | 5.15 | 0.2 | 0.6 | 1.2 | 3.9 | 0.09 | 0.2 | 0.11 | 1.0 | 0.7 | 0.2 | 0.3 | 0.1 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 5 |
| traffi | 26/1 | 9.0 | 4.76 | 2.99 | 0.1 | 0.4 | 0.4 | 1.3 | 0.07 | 0.0 | 0.00 | 0.3 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 7 |
|  | 28/1 | 1.4 | 0.36 | 0.20 | 0.0 | 0.0 | 0.0 | 0.0 | 0.07 | 0.0 | 0.00 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
|  | 2/2/ | 11. | 2.75 | 1.70 | 0.0 | 0.3 | 0.3 | 1.2 | 0.07 | 0.0 | 0.00 | 0.3 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 1 |

| | | | | | | | | | | | | | | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 4/2/ | 7.4 | 2.30 | 1.48 | 0.1 | 0.1 | 0.2 | 0.9 | 0.07 | 0.0 | 0.00 | 0.1 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 1 |
| | 9/2/ | 16. | 3.82 | 2.44 | 0.1 | 0.3 | 0.4 | 1.6 | 0.09 | 0.0 | 0.00 | 0.4 | 0.2 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 2 |
| | 11/2 | 4.9 | 1.52 | 0.94 | 0.1 | 0.1 | 0.0 | 0.4 | 0.10 | 0.0 | 0.00 | 0.0 | 0.0 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.1 | 0.3 | 0.1 | 4 |
| | 16/2 | 27. | 8.03 | 5.06 | 0.1 | 1.2 | 1.6 | 4.4 | 0.11 | 0.3 | 0.08 | 2.3 | 1.4 | 1.0 | 1.3 | 0.2 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 6 |
| M06<br>traffi | 21/1 | 7.3 | 1.62 | 1.06 | 0.8 | 0.1 | 0.1 | 0.7 | 0.09 | 0.1 | 0.00 | 0.2 | 0.1 | 0.1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.1 | 0.2 | 0.3 | 0.1 | 7 |
| | 26/1 | Technical problem PAH measurment | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 28/1 | | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 2/2/ | | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 4/2/ | | | | | | | | | | | | | | | | | | | | | | | 1 |
| | 9/2/ | | | | | | | | | | | | | | | | | | | | | | | 2 |
| | 11/2 | | | | | | | | | | | | | | | | | | | | | | | 5 |
| | 16/2 | | | | | | | | | | | | | | | | | | | | | | | 6 |

Table 21).

*In summary*

On the traffic and the industry locations of Genk/Diepenbeek, the concentrations of lower molecular weight PAHs such as NAP (and methylated forms), ACY, ACE and ANT were ca. 3-7 times higher than the levels at the background location in that area. On the traffic location, also FLU, FLA, PYR and BghiP were in the same way relatively higher, mainly in summer. These are compounds typical for traffic emissions. In Menen, most PAHs were homogeneously appearing over the whole area (industry, traffic and background locations). Only in summer-fall the levels of ANT, BkF and IcdP were relatively 3-6 times higher on the industry locations of Menen, compared to those at the background location of Menen.

*Table 16 PAHs and levoglucosan (LEV) levels in Genk/Diepenbeek during the summer-fall (Sept'15) campaign given in ng/m$^3$. Gradual color scale of which green indicates the lowest and red the largest value. The color scale was applied to each PAH individually but over all areas and all seasons. Each line is an individual sample for which the column 'start-PAH' gives the start time of the 48h-sampling*

| ID | start-PAK | NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP | LEV |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| G01 | 1/9/15 9:30 AM | 46.488 | 40.172 | 20.472 | 0.674 | 12.762 | 11.237 | 19.811 | 1.326 | 1.279 | 0.796 | 2.842 | 3.024 | 1.094 | 0.529 | 1.239 | 0.724 | 1.150 | 1.290 | 6.935 | 8.349 | 10.316 | 6.593 | 20 |
| traffic | 3/9/15 9:55 AM | 33.792 | 13.410 | 5.506 | 0.198 | 3.814 | 5.602 | 13.362 | 1.115 | 0.646 | 0.526 | 2.005 | 2.424 | 1.239 | 0.523 | 0.952 | 0.515 | 0.680 | 0.795 | 3.124 | 3.299 | 4.259 | 3.045 | 23 |
|  | 8/9/15 9:0 AM | 37.603 | 16.582 | 9.648 | 2.018 | 3.494 | 4.391 | 10.969 | 1.146 | 0.645 | 0.722 | 2.427 | 2.949 | 1.123 | 0.637 | 1.203 | 0.627 | 0.735 | 0.931 | 2.092 | 2.115 | 3.385 | 2.190 | 31 |
|  | 17/9/15 9:10 AM | 18.136 | 6.104 | 2.976 | 0.082 | 1.590 | 2.805 | 7.526 | 0.799 | 0.514 | 0.458 | 1.928 | 1.996 | 0.998 | 0.628 | 1.016 | 0.590 | 0.682 | 0.840 | 1.782 | 1.654 | 2.300 | 1.482 |  |
|  | 22/9/15 9:15 AM | 12.327 | 5.409 | 3.097 | 0.098 | 1.301 | 2.228 | 6.843 | 0.893 | 0.005 | 0.293 | 3.478 | 5.919 | 0.955 | 0.712 | 1.093 | 0.594 | 1.500 | 0.915 | 1.304 | 1.195 | 1.479 | 1.628 | 64 |
|  | 24/9/15 9:45 AM | 13.878 | 4.903 | 2.589 | 0.065 | 1.217 | 2.326 | 6.336 | 0.812 | 0.375 | 0.392 | 1.501 | 2.181 | 1.234 | 0.497 | 0.876 | 0.532 | 0.673 | 0.686 | 0.953 | 1.016 | 1.113 | 0.881 | 128 |
|  | 29/9/15 8:50 AM | 42.792 | 9.817 | 5.692 | 2.737 | 1.410 | 2.763 | 6.895 | 0.909 | 0.576 | 0.466 | 2.053 | 2.700 | 1.099 | 0.669 | 1.180 | 0.591 | 0.718 | 0.822 | 1.370 | 1.014 | 1.111 | 0.845 | 32 |
|  | 1/10/15 9:15 AM | 27.656 | 7.718 | 3.691 | 2.058 | 0.768 | 1.931 | 4.813 | 0.893 | 0.266 | 0.412 | 2.093 | 2.597 | 1.094 | 0.744 | 1.213 | 0.633 | 0.728 | 0.888 | 1.127 | 1.008 | 0.842 | 0.755 | 208 |
| G02 | 1/9/15 9:30 AM | 23.883 | 11.993 | 6.625 | 0.678 | 2.718 | 3.498 | 8.063 | 0.929 | 1.502 | 1.628 | 2.528 | 2.459 | 3.389 | 3.466 | 5.283 | 5.570 | 5.036 | 6.992 | 11.468 | 14.706 | 20.450 | 9.324 |  |
| traffic | 3/9/15 9:25 AM | 9.213 | 2.480 | 1.450 | 0.167 | 0.675 | 0.838 | 2.344 | 0.317 | 0.470 | 0.458 | 1.035 | 1.016 | 1.430 | 1.637 | 2.103 | 2.493 | 2.194 | 2.778 | 3.801 | 4.620 | 6.024 | 3.248 |  |
|  | 8/9/15 9:30 AM | 44.787 | 17.683 | 10.610 | 0.473 | 2.612 | 3.795 | 8.725 | 0.179 | 0.960 | 0.540 | 1.950 | 1.625 | 1.531 | 1.647 | 2.134 | 2.219 | 1.978 | 2.655 | 4.434 | 5.458 | 8.169 | 3.983 |  |
|  | 10/9/15 9:35 AM | 6.121 | 1.505 | 0.587 | 0.256 | 0.176 | 0.347 | 1.607 | 1.825 | 0.352 | 1.020 | 5.011 | 7.754 | 2.173 | 2.259 | 2.737 | 1.741 | 4.363 | 2.163 | 1.840 | 1.219 | 0.886 | 6.491 |  |
|  | 14/9/15 10:0 AM | 18.964 | 9.790 | 6.527 | 1.323 | 13.801 | 9.383 | 6.102 | 0.547 | 0.587 | 0.201 | 1.572 | 1.943 | 0.878 | 0.401 | 0.662 | 0.314 | 0.520 | 0.501 | 0.852 | 0.608 | 0.719 | 1.414 |  |
|  | 19/9/15 9:35 AM | 65.729 | 23.353 | 13.317 | 0.467 | 3.423 | 5.442 | 12.385 | 1.630 | 0.371 | 0.756 | 4.825 | 4.524 | 1.306 | 1.031 | 1.817 | 0.702 | 1.019 | 1.132 | 1.714 | 1.330 | 1.029 | 1.374 |  |
|  | 22/9/15 9:30 AM | 53.100 | 20.811 | 9.693 | 0.611 | 4.903 | 7.302 | 17.135 | 1.386 | 0.532 | 0.788 | 5.881 | 6.122 | 1.427 | 1.030 | 1.694 | 0.699 | 1.139 | 1.150 | 1.274 | 0.892 | 1.182 | 1.256 |  |
|  | 24/9/15 9:30 AM | 25.461 | 11.265 | 5.035 | 1.101 | 2.823 | 4.600 | 10.441 | 0.997 | 0.005 | 0.547 | 4.507 | 5.230 | 1.371 | 0.703 | 1.295 | 0.515 | 1.236 | 0.933 | 1.720 | 1.100 | 0.960 | 1.499 |  |
| G03 | 1/9/15 7:0 AM | 0.025 | 0.005 | 0.005 | 0.005 | 0.005 | 0.006 | 0.048 | 0.005 | 0.005 | 0.038 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.125 | 0.005 | 0.005 | 0.125 | 19 |
| industry | 3/9/15 7:15 AM | 0.480 | 0.105 | 0.005 | 0.052 | 0.005 | 0.073 | 0.354 | 0.066 | 0.148 | 0.121 | 0.270 | 0.254 | 0.353 | 0.473 | 0.324 | 0.356 | 0.288 | 0.342 | 0.456 | 0.514 | 0.637 | 0.471 | 27 |
|  | 8/9/15 7:15 AM | 0.572 | 0.062 | 0.005 | 0.049 | 0.005 | 0.053 | 0.316 | 0.112 | 0.117 | 0.108 | 0.308 | 0.243 | 0.352 | 0.475 | 0.333 | 0.410 | 0.271 | 0.317 | 0.412 | 0.473 | 0.565 | 0.431 | 26 |
|  | 10/9/15 7:20 AM | 1.554 | 1.059 | 0.268 | 0.005 | 0.005 | 0.069 | 0.733 | 0.516 | 0.005 | 0.171 | 0.502 | 0.502 | 0.736 | 0.381 | 1.002 | 0.455 | 0.541 | 0.721 | 0.677 | 0.709 | 0.480 | 0.935 | 37 |
|  | 15/9/15 7:5 AM | 39.369 | 9.313 | 6.352 | 0.174 | 2.977 | 3.566 | 8.659 | 0.894 | 0.285 | 0.437 | 2.077 | 2.393 | 1.232 | 0.513 | 1.098 | 0.489 | 0.642 | 0.658 | 1.085 | 1.039 | 0.492 | 0.742 | 31 |
|  | 17/9/15 7:15 AM | 8.694 | 2.335 | 1.074 | 0.029 | 0.492 | 0.792 | 2.269 | 0.584 | 0.155 | 0.220 | 0.690 | 0.903 | 1.345 | 0.379 | 0.710 | 0.423 | 0.530 | 0.617 | 0.558 | 0.681 | 0.005 | 0.276 | 12 |
|  | 22/9/15 7:15 AM | 67.460 | 21.554 | 10.037 | 0.816 | 3.665 | 5.380 | 14.294 | 1.176 | 0.562 | 0.704 | 2.959 | 2.159 | 1.214 | 0.746 | 1.290 | 0.526 | 0.754 | 0.917 | 0.878 | 0.721 | 0.556 | 0.367 | 68 |
|  | 24/9/15 7:20 AM | 5.443 | 1.319 | 0.576 | 0.005 | 0.125 | 0.381 | 1.423 | 0.542 | 0.152 | 0.215 | 0.618 | 0.534 | 0.676 | 0.382 | 0.736 | 0.419 | 0.533 | 0.670 | 0.570 | 0.675 | 0.330 | 0.125 | 66 |
| G04 | 1/9/15 8:30 AM | 8.025 | 1.097 | 0.597 | 1.175 | 5.565 | 0.158 | 0.576 | 0.129 | 0.252 | 0.129 | 0.437 | 0.469 | 0.572 | 0.767 | 0.807 | 0.841 | 0.721 | 0.946 | 0.877 | 1.076 | 1.443 | 1.013 |  |
| industry | 3/9/15 8:30 AM | 3.695 | 0.433 | 0.259 | 0.991 | 3.422 | 0.126 | 0.559 | 0.085 | 0.225 | 0.129 | 0.417 | 0.338 | 0.426 | 0.628 | 0.558 | 0.655 | 0.572 | 0.667 | 0.672 | 0.689 | 0.772 | 0.577 |  |
|  | 8/9/15 8:20 AM | 12.555 | 1.828 | 0.925 | 1.111 | 7.311 | 0.149 | 0.546 | 0.070 | 0.246 | 0.125 | 0.364 | 0.386 | 0.551 | 0.702 | 0.689 | 0.780 | 0.679 | 0.948 | 0.896 | 1.125 | 1.509 | 0.823 |  |
|  | 10/9/15 8:15 AM | 12.990 | 1.833 | 0.780 | 1.532 | 11.072 | 0.105 | 0.549 | 0.005 | 0.124 | 0.247 | 0.300 | 0.344 | 0.614 | 0.333 | 0.684 | 0.411 | 0.508 | 0.587 | 0.549 | 0.728 | 0.005 | 0.125 |  |
|  | 15/9/15 10:45 AM | 16.152 | 2.119 | 0.546 | 1.246 | 18.124 | 0.005 | 0.220 | 0.472 | 0.081 | 0.262 | 0.271 | 0.270 | 0.866 | 0.701 | 1.016 | 0.728 | 0.890 | 1.174 | 2.243 | 2.509 | 2.556 | 1.988 |  |
|  | 17/9/15 10:15 AM | 4.301 | 0.739 | 0.159 | 1.195 | 6.471 | 0.005 | 0.115 | 0.005 | 0.027 | 0.190 | 0.206 | 0.170 | 0.659 | 0.415 | 0.731 | 0.479 | 0.631 | 0.814 | 1.118 | 1.281 | 1.022 | 0.898 |  |
|  | 23/9/15 8:30 AM | 10.397 | 1.582 | 0.570 | 1.323 | 12.049 | 0.005 | 0.206 | 0.005 | 0.089 | 0.186 | 0.212 | 0.205 | 0.645 | 0.398 | 0.743 | 0.443 | 0.579 | 0.756 | 1.048 | 1.234 | 1.197 | 0.679 | 35 |
|  | 25/9/15 8:10 AM | 6.202 | 1.131 | 0.294 | 1.159 | 6.610 | 0.005 | 0.137 | 0.440 | 0.034 | 0.187 | 0.185 | 0.190 | 0.599 | 0.350 | 0.670 | 0.360 | 0.528 | 0.675 | 0.733 | 0.932 | 0.766 | 0.357 | 52 |
| G05 | 31/8/15 2:5 PM | 4.919 | 3.195 | 1.700 | 0.129 | 0.998 | 1.918 | 5.534 | 0.035 | 0.005 | 0.166 | 0.698 | 0.396 | 0.294 | 0.340 | 0.407 | 0.425 | 0.328 | 0.405 | 0.642 | 0.781 | 1.268 | 0.522 | 19 |
| backgrnd | 4/9/15 3:30 PM | 10.779 | 5.457 | 3.241 | 1.110 | 1.627 | 2.173 | 5.999 | 0.083 | 0.005 | 0.248 | 0.949 | 0.801 | 0.527 | 0.593 | 0.708 | 0.732 | 0.557 | 0.681 | 0.952 | 1.247 | 1.916 | 0.798 | 40 |
|  | 8/9/15 1:0 PM | 30.685 | 9.310 | 5.831 | 0.165 | 1.634 | 2.439 | 5.614 | 0.069 | 0.005 | 0.231 | 0.899 | 0.792 | 0.507 | 0.643 | 0.795 | 0.668 | 0.591 | 0.679 | 0.932 | 1.117 | 1.678 | 0.803 | 23 |
|  | 10/9/15 1:32 PM | 8.419 | 1.758 | 0.756 | 0.005 | 0.109 | 0.181 | 0.634 | 0.460 | 0.026 | 0.166 | 0.386 | 0.535 | 0.616 | 0.361 | 0.698 | 0.371 | 0.586 | 0.654 | 0.733 | 0.880 | 0.784 | 0.335 | 35 |
|  | 15/9/15 10:0 AM | 6.450 | 1.272 | 0.412 | 0.005 | 0.132 | 0.178 | 0.883 | 0.501 | 0.066 | 0.183 | 0.450 | 0.483 | 0.737 | 0.380 | 0.726 | 0.364 | 0.567 | 0.679 | 0.736 | 0.889 | 0.689 | 0.261 | 50 |
|  | 17/9/15 7:50 AM |  |  |  |  |  |  |  |  |  | no PAH measurement |  |  |  |  |  |  |  |  |  |  |  |  | 33 |
|  | 22/9/15 9:30 AM | 23.695 | 2.489 | 1.707 | 0.254 | 0.548 | 0.779 | 2.612 | 0.513 | 0.147 | 0.256 | 0.754 | 0.475 | 0.793 | 0.327 | 0.706 | 0.319 | 0.494 | 0.604 | 0.487 | 0.607 | 0.325 | 0.125 | 86 |
|  | 24/9/15 9:32 AM | 21.700 | 3.463 | 1.755 | 0.102 | 0.702 | 0.735 | 2.290 | 0.527 | 0.118 | 0.219 | 0.701 | 0.429 | 0.632 | 0.402 | 0.694 | 0.313 | 0.495 | 0.592 | 0.524 | 0.616 | 0.293 | 0.125 | 102 |
| G06 | 2/9/15 10:20 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 32 |
| backgrnd | 3/9/15 10:10 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 16 |
|  | 8/9/15 10:40 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 23 |
|  | 10/9/15 5:30 PM |  |  |  |  |  |  |  |  |  | Technical problems PAH measurement |  |  |  |  |  |  |  |  |  |  |  |  | 35 |
|  | 15/9/15 10:0 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 17 |
|  | 17/9/15 10:0 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 30 |
|  | 21/9/15 11:0 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 36 |
|  | 24/9/15 6:0 PM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 119 |

*Table 17 PAHs and levoglucosan(LEV) levels in Menen during the summer-fall (Sept'15) campaign given in ng/m$^3$. Gradual color scale of which green indicates the lowest and red the largest value. The color scale was applied to each PAH individually but over all areas and all seasons. Each line is an individual sample for which the column 'start-PAH' gives the start time of the 48h-sampling*

| ID | start-PAK | NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP | LEV |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| M01<br>industry | 1/9/15 8:11 AM | 21.435 | 15.413 | 8.562 | 0.207 | 2.603 | 3.377 | 7.897 | 0.035 | 0.005 | 0.193 | 1.354 | 0.769 | 0.525 | 0.850 | 0.636 | 0.562 | 0.452 | 0.479 | 0.805 | 0.908 | 1.295 | 0.666 | 14 |
|  | 3/9/15 8:59 AM | 23.308 | 11.085 | 6.958 | 0.187 | 1.870 | 3.319 | 7.445 | 0.026 | 0.005 | 0.217 | 1.424 | 1.074 | 0.502 | 0.920 | 0.747 | 0.542 | 0.466 | 0.498 | 0.785 | 0.898 | 1.174 | 0.623 | 22 |
|  | 8/9/15 8:27 AM | 23.152 | 14.866 | 8.979 | 0.439 | 2.622 | 4.431 | 9.071 | 0.038 | 0.005 | 0.266 | 1.597 | 0.927 | 0.418 | 0.636 | 0.649 | 0.493 | 0.486 | 0.355 | 0.659 | 0.721 | 1.054 | 0.589 | 42 |
|  | 10/9/15 8:56 AM | 32.360 | 21.539 | 12.126 | 0.376 | 3.299 | 6.133 | 11.162 | 0.072 | 0.005 | 0.327 | 2.563 | 1.490 | 0.430 | 0.739 | 0.727 | 0.607 | 0.477 | 0.550 | 0.720 | 0.744 | 1.132 | 0.612 | 39 |
|  | 15/9/15 8:14 AM | 24.199 | 3.580 | 1.629 | 0.005 | 0.324 | 0.574 | 2.070 | 0.521 | 0.125 | 0.231 | 0.762 | 0.481 | 0.654 | 0.375 | 0.703 | 0.299 | 0.479 | 0.570 | 0.469 | 0.622 | 0.005 | 0.125 | 100 |
|  | 17/9/15 8:35 AM | 19.131 | 3.283 | 1.504 | 0.005 | 0.361 | 0.596 | 2.120 | 0.524 | 0.066 | 0.228 | 0.815 | 0.503 | 0.638 | 0.390 | 0.677 | 0.299 | 0.474 | 0.564 | 0.483 | 0.601 | 0.005 | 0.125 | 70 |
|  | 22/9/15 7:3 AM | 19.724 | 0.473 | 1.273 | 0.005 | 0.260 | 0.472 | 1.555 | 0.501 | 0.080 | 0.214 | 0.597 | 0.445 | 0.570 | 0.509 | 0.652 | 0.280 | 0.452 | 0.568 | 0.443 | 0.577 | 0.005 | 0.125 | 52 |
|  | 24/9/15 10:8 AM | 54.253 | 8.953 | 4.008 | 0.028 | 0.772 | 0.949 | 3.066 | 0.568 | 0.186 | 0.284 | 1.054 | 0.698 | 0.692 | 0.477 | 0.846 | 0.339 | 0.530 | 0.632 | 0.510 | 0.601 | 0.005 | 0.125 | 169 |
| M02<br>industry | 1/9/15 7:30 AM | 16.861 | 11.072 | 6.484 | 0.371 | 1.611 | 2.238 | 4.467 | 0.060 | 0.005 | 0.223 | 0.822 | 0.683 | 0.389 | 0.441 | 0.471 | 0.424 | 0.326 | 0.379 | 0.447 | 0.539 | 0.740 | 0.397 | 27 |
|  | 3/9/15 7:35 AM | 18.347 | 10.113 | 5.880 | 0.214 | 1.346 | 2.211 | 4.322 | 0.052 | 0.005 | 0.197 | 0.758 | 0.642 | 0.301 | 0.401 | 0.446 | 0.346 | 0.300 | 0.349 | 0.458 | 0.508 | 0.723 | 0.400 | 72 |
|  | 8/9/15 7:45 AM | 8.499 | 4.985 | 2.955 | 0.104 | 0.998 | 1.539 | 2.572 | 0.038 | 0.005 | 0.123 | 0.555 | 0.471 | 0.232 | 0.305 | 0.338 | 0.311 | 0.239 | 0.262 | 0.334 | 0.376 | 0.558 | 0.296 | 28 |
|  | 10/9/15 8:0 AM | 20.514 | 13.722 | 7.998 | 0.252 | 1.760 | 3.864 | 6.652 | 0.091 | 0.005 | 0.277 | 1.188 | 0.920 | 0.305 | 0.476 | 0.449 | 0.358 | 0.288 | 0.286 | 0.444 | 0.481 | 0.683 | 0.392 | 55 |
|  | 15/9/15 7:15 AM | 13.141 | 2.336 | 1.071 | 0.005 | 0.096 | 0.252 | 0.930 | 0.479 | 0.117 | 0.193 | 0.441 | 0.316 | 0.573 | 0.431 | 0.637 | 0.289 | 0.464 | 0.545 | 0.472 | 0.613 | 0.005 | 0.125 | 31 |
|  | 17/9/15 7:45 AM | 13.077 | 2.358 | 1.031 | 0.005 | 0.104 | 0.203 | 0.788 | 0.474 | 0.064 | 0.185 | 0.336 | 0.288 | 0.743 | 0.296 | 0.626 | 0.290 | 0.455 | 0.552 | 0.466 | 0.005 | 0.005 | 0.125 | 17 |
|  | 22/9/15 7:30 AM | 13.063 | 2.295 | 1.019 | 0.005 | 0.121 | 0.236 | 0.888 | 0.470 | 0.054 | 0.185 | 0.399 | 0.337 | 0.583 | 0.389 | 0.660 | 0.297 | 0.468 | 0.552 | 0.473 | 0.594 | 0.005 | 0.125 | 111 |
|  | 24/9/15 7:45 AM | 19.988 | 3.997 | 1.838 | 0.191 | 0.395 | 0.275 | 0.781 | 0.458 | 0.066 | 0.184 | 0.335 | 0.256 | 0.577 | 0.336 | 0.659 | 0.300 | 0.475 | 0.582 | 0.461 | 0.005 | 0.005 | 0.125 | 204 |
| M03<br>backgrnd | 1/9/15 7:25 AM | 15.101 | 11.831 | 7.154 | 0.185 | 1.283 | 2.301 | 5.563 | 0.046 | 0.005 | 0.214 | 1.281 | 1.389 | 0.378 | 0.425 | 0.463 | 0.324 | 0.416 | 0.333 | 0.436 | 0.475 | 0.640 | 0.554 | 14 |
|  | 3/9/15 7:30 AM | 13.574 | 11.101 | 7.301 | 0.185 | 1.333 | 2.598 | 4.028 | 0.006 | 0.005 | 0.172 | 0.703 | 0.574 | 0.226 | 0.330 | 0.347 | 0.266 | 0.223 | 0.245 | 0.344 | 0.359 | 0.557 | 0.359 | 19 |
|  | 8/9/15 7:30 AM | 17.069 | 10.981 | 5.884 | 0.177 | 1.737 | 2.886 | 5.425 | 0.005 | 0.005 | 0.178 | 0.877 | 0.614 | 0.177 | 0.314 | 0.288 | 0.239 | 0.189 | 0.179 | 0.306 | 0.291 | 0.517 | 0.392 | 24 |
|  | 10/9/15 7:30 AM | 20.797 | 12.710 | 7.273 | 0.195 | 1.268 | 3.274 | 4.725 | 0.040 | 0.005 | 0.156 | 0.724 | 0.569 | 0.183 | 0.291 | 0.292 | 0.215 | 0.177 | 0.199 | 0.278 | 0.281 | 0.462 | 0.287 | 35 |
|  | 15/9/15 7:30 AM | 38.506 | 10.750 | 7.547 | 0.005 | 1.219 | 2.819 | 7.882 | 0.497 | 0.350 | 0.415 | 1.273 | 2.252 | 1.112 | 0.005 | 0.531 | 0.005 | 0.382 | 0.201 | 0.125 | 0.005 | 0.005 | 0.125 | 33 |
|  | 17/9/15 7:30 AM | 52.926 | 15.991 | 12.894 | 0.193 | 1.688 | 3.876 | 9.388 | 0.546 | 0.857 | 0.499 | 1.348 | 1.249 | 0.655 | 0.368 | 0.574 | 0.005 | 0.161 | 0.202 | 0.125 | 0.005 | 0.005 | 0.125 | 68 |
|  | 22/9/15 7:15 AM | 42.964 | 20.477 | 13.690 | 0.188 | 2.600 | 4.686 | 8.414 | 0.922 | 0.005 | 0.531 | 1.329 | 1.192 | 0.436 | 0.449 | 0.570 | 0.005 | 0.166 | 0.247 | 0.125 | 0.005 | 0.005 | 0.125 | 31 |
|  | 24/9/15 7:30 AM | 67.477 | 29.149 | 19.272 | 1.802 | 3.315 | 4.747 | 9.293 | 0.631 | 0.067 | 0.667 | 1.797 | 1.098 | 0.426 | 0.702 | 0.929 | 0.029 | 0.365 | 0.510 | 0.125 | 0.005 | 0.005 | 0.125 | 181 |
| M04<br>backgrnd | 1/9/15 8:25 AM | 12.897 | 6.796 | 3.156 | 0.092 | 0.652 | 1.395 | 2.594 | 0.033 | 0.005 | 0.152 | 0.517 | 0.402 | 0.151 | 0.268 | 0.229 | 0.173 | 0.137 | 0.138 | 0.266 | 0.256 | 0.379 | 0.125 |  |
|  | 3/9/15 12:55 PM | 12.653 | 5.297 | 3.643 | 0.080 | 0.512 | 1.211 | 1.733 | 0.005 | 0.005 | 0.087 | 0.378 | 0.319 | 0.153 | 0.218 | 0.195 | 0.128 | 0.118 | 0.128 | 0.125 | 0.220 | 0.321 | 0.125 |  |
|  | 8/9/15 9:10 AM | 12.419 | 5.491 | 3.077 | 0.279 | 0.672 | 1.378 | 2.539 | 0.005 | 0.005 | 0.125 | 0.488 | 0.591 | 0.113 | 0.172 | 0.144 | 0.120 | 0.131 | 0.101 | 0.125 | 0.169 | 0.277 | 0.292 |  |
|  | 10/9/15 9:17 AM | 13.551 | 5.402 | 3.178 | 0.074 | 0.531 | 1.547 | 2.251 | 0.005 | 0.005 | 0.079 | 0.314 | 0.288 | 0.103 | 0.136 | 0.104 | 0.081 | 0.063 | 0.068 | 0.125 | 0.141 | 0.203 | 0.125 |  |
|  | 15/9/15 9:0 AM | 30.811 | 9.476 | 5.355 | 0.005 | 0.714 | 2.301 | 6.194 | 0.432 | 0.005 | 0.344 | 0.993 | 1.098 | 0.576 | 0.233 | 0.427 | 0.005 | 0.086 | 0.140 | 0.125 | 0.005 | 0.005 | 0.125 |  |
|  | 17/9/15 9:15 AM | 43.341 | 12.930 | 8.067 | 0.065 | 1.105 | 2.901 | 6.891 | 0.483 | 0.005 | 0.413 | 1.150 | 0.978 | 0.421 | 0.256 | 0.449 | 0.005 | 0.098 | 0.185 | 0.125 | 0.234 | 0.005 | 0.125 |  |
|  | 22/9/15 11:40 AM | 42.140 | 13.555 | 9.194 | 0.123 | 1.667 | 3.359 | 7.187 | 0.572 | 0.005 | 0.460 | 1.276 | 1.134 | 0.540 | 0.213 | 0.494 | 0.005 | 0.122 | 0.217 | 0.125 | 0.262 | 0.005 | 0.125 | 47 |
|  | 24/9/15 11:45 AM | 66.590 | 23.830 | 16.875 | 0.508 | 3.441 | 4.510 | 8.727 | 0.609 | 0.005 | 0.480 | 1.714 | 1.136 | 0.449 | 0.405 | 0.698 | 0.005 | 0.239 | 0.360 | 0.125 | 0.243 | 0.005 | 0.125 | 130 |
| M05<br>traffic | 1/9/15 9:45 AM | 10.196 | 4.565 | 2.570 | 0.005 | 0.983 | 1.222 | 2.282 | 0.035 | 0.005 | 0.121 | 0.320 | 0.198 | 0.368 | 0.149 | 0.092 | 0.073 | 0.051 | 0.058 | 0.125 | 0.121 | 0.158 | 0.125 | 20 |
|  | 3/9/15 9:15 AM | 10.905 | 3.033 | 1.330 | 0.034 | 0.416 | 0.665 | 1.063 | 0.078 | 0.005 | 0.066 | 0.196 | 0.133 | 0.077 | 0.200 | 0.068 | 0.052 | 0.035 | 0.033 | 0.125 | 0.084 | 0.137 | 0.125 | 49 |
|  | 8/9/15 9:40 AM | 13.796 | 4.611 | 2.103 | 0.056 | 1.105 | 1.251 | 2.042 | 0.083 | 0.005 | 0.060 | 0.278 | 0.143 | 0.078 | 0.183 | 0.054 | 0.038 | 0.027 | 0.029 | 0.125 | 0.079 | 0.100 | 0.125 | 28 |
|  | 26/9/15 11:30 AM | 91.594 | 29.765 | 18.362 | 0.834 | 12.500 | 12.515 | 23.286 | 0.908 | 1.496 | 0.968 | 3.547 | 2.001 | 0.728 | 0.548 | 1.047 | 0.047 | 0.369 | 0.492 | 0.125 | 0.242 | 0.337 | 0.125 | 53 |
|  | 17/9/15 11:15 AM | 46.582 | 17.470 | 11.494 | 0.212 | 2.171 | 4.051 | 9.945 | 0.699 | 0.005 | 0.465 | 1.828 | 1.266 | 0.577 | 0.395 | 0.653 | 0.005 | 0.175 | 0.257 | 0.125 | 0.274 | 0.509 | 0.125 | 51 |
|  | 15/9/15 10:0 AM | 33.149 | 15.626 | 9.767 | 0.130 | 2.541 | 4.373 | 10.704 | 0.873 | 0.602 | 0.533 | 1.931 | 1.556 | 0.423 | 0.421 | 0.607 | 0.005 | 0.174 | 0.249 | 0.125 | 0.274 | 0.402 | 0.125 | 47 |
|  | 22/9/15 10:0 AM | 37.477 | 18.641 | 13.306 | 0.173 | 3.036 | 4.556 | 9.070 | 0.233 | 0.354 | 0.461 | 2.491 | 2.684 | 0.924 | 1.095 | 1.235 | 0.300 | 1.568 | 0.869 | 0.125 | 0.005 | 0.628 | 0.125 | 40 |
|  | 24/9/15 11:15 AM | 76.310 | 36.419 | 26.469 | 1.915 | 7.006 | 7.025 | 13.247 | 0.956 | 0.391 | 0.714 | 2.456 | 1.692 | 0.681 | 0.717 | 1.067 | 0.072 | 0.550 | 0.731 | 0.125 | 0.268 | 0.482 | 0.125 | 183 |
| M06<br>traffic | 1/9/15 7:45 AM | 1.620 | 0.825 | 0.197 | 0.005 | 0.005 | 0.005 | 0.263 | 0.433 | 0.023 | 0.156 | 0.213 | 0.194 | 0.543 | 0.280 | 0.584 | 0.280 | 0.440 | 0.566 | 0.450 | 0.005 | 0.005 | 0.125 | 24 |
|  | 3/9/15 8:30 AM | 0.036 | 0.475 | 0.005 | 0.005 | 0.005 | 0.005 | 0.080 | 0.416 | 0.005 | 0.151 | 0.167 | 0.136 | 0.527 | 0.269 | 0.005 | 0.275 | 0.430 | 0.538 | 0.125 | 0.569 | 0.005 | 0.125 | 42 |
|  | 8/9/15 8:0 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 382 |
|  | 10/9/15 10:5 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 129 |
|  | 15/9/15 8:0 AM |  |  |  |  |  |  |  |  |  | no PAH measurements |  |  |  |  |  |  |  |  |  |  |  |  | 39 |
|  | 17/9/15 8:10 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 70 |
|  | 22/9/15 7:45 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 42 |
|  | 24/9/15 10:20 AM |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  | 152 |

*Table 18 PAHs and levoglucosan(LEV) levels in Houtem during the summer-fall (Sept'15) campaign given in ng/m$^3$. Gradual color scale of which green indicates the lowest and red the largest value. The color scale was applied to each PAH individually but over all areas and all seasons. Each line is an individual sample for which the column 'start-PAH' gives the start time of the 48h-sampling*

| ID | start-PAK | NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP | LEV |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| H backgrnd | 2/9/15 2:41 PM | 0.875 | 0.168 | 0.098 | 0.005 | 0.013 | 0.056 | 0.110 | 0.023 | 0.025 | 0.005 | 0.071 | 0.101 | 0.036 | 0.067 | 0.042 | 0.036 | 0.034 | 0.025 | 0.125 | 0.059 | 0.081 | 0.125 | 19 |
| | 3/9/15 5:47 PM | 0.577 | 1.267 | 0.642 | 0.005 | 0.028 | 0.060 | 0.068 | 0.005 | 0.013 | 0.012 | 0.015 | 0.017 | 0.012 | 0.020 | 0.017 | 0.017 | 0.005 | 0.005 | 0.125 | 0.017 | 0.032 | 0.125 | 15 |
| | 8/9/15 6:7 PM | 3.989 | 0.378 | 0.216 | 0.005 | 0.005 | 0.072 | 0.102 | 0.005 | 0.005 | 0.005 | 0.016 | 0.016 | 0.025 | 0.033 | 0.018 | 0.015 | 0.011 | 0.013 | 0.125 | 0.031 | 0.051 | 0.125 | 13 |
| | 10/9/15 6:24 PM | 0.353 | 0.128 | 0.090 | 0.005 | 0.005 | 0.032 | 0.042 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.125 | 0.005 | 0.013 | 0.125 | 24 |
| | 15/9/15 12:1 AM | no PAH measurement | | | | | | | | | | | | | | | | | | | | | | 36 |
| | 17/9/15 5:36 PM | 1.771 | 0.596 | 0.175 | 0.005 | 0.005 | 0.005 | 0.322 | 0.106 | 0.005 | 0.020 | 0.005 | 0.005 | 0.083 | 0.022 | 0.161 | 0.005 | 0.005 | 0.078 | 0.125 | 0.005 | 0.005 | 0.125 | 24 |
| | 22/9/15 6:41 PM | 4.166 | 0.970 | 0.448 | 0.005 | 0.005 | 0.176 | 0.442 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.090 | 0.033 | 0.184 | 0.005 | 0.005 | 0.144 | 0.125 | 0.005 | 0.005 | 0.125 | 21 |
| | 25/9/15 8:45 AM | 6.076 | 1.158 | 0.427 | 0.005 | 0.005 | 0.270 | 0.885 | 0.190 | 0.005 | 0.050 | 0.005 | 0.005 | 0.179 | 0.085 | 0.308 | 0.005 | 0.013 | 0.130 | 0.125 | 0.005 | 0.005 | 0.125 | 75 |

*Table 19 PAHs and levoglucosan(LEV) levels in Genk/Diepenbeek during the winter (Jan-Feb) campaign, given in ng/m$^3$. Gradual color scale of which green indicates the lowest and red the largest value. The color scale was applied to each PAH individually but over all areas and all seasons. Each line is an individual sample for which the column 'start-PAH' gives the start time of the 48h-sampling*

| ID | start-PAK | NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP | LEV |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| G21 | 19/1/16 11:30 AM | 70.649 | 30.208 | 22.017 | 4.266 | 5.721 | 10.238 | 28.102 | 0.333 | 2.999 | 0.921 | 11.377 | 7.183 | 2.223 | 3.387 | 2.043 | 0.630 | 0.562 | 0.778 | 0.125 | 0.159 | 0.252 | 0.125 | 938 |
| traffic | 24/1/16 10:50 AM | 32.770 | 8.929 | 5.746 | 0.219 | 2.175 | 3.640 | 9.462 | 0.079 | 0.228 | 0.224 | 2.791 | 1.513 | 0.301 | 0.530 | 0.511 | 0.136 | 0.185 | 0.222 | 0.125 | 0.169 | 0.253 | 0.125 | 219 |
| | 30/1/16 3:30 PM | | | | | | | | | | | | | | | | | | | | | | | 73 |
| | 12/2/16 2:15 PM | | | | | | | | | | Technical problems PAH measurement | | | | | | | | | | | | | 290 |
| | 16/2/16 5:0 PM | | | | | | | | | | | | | | | | | | | | | | | 362 |
| G29 | 19/1/16 8:30 AM | 87.570 | 33.235 | 23.296 | 8.949 | 5.849 | 12.117 | 32.640 | 0.143 | 2.411 | 1.097 | 13.727 | 7.811 | 0.005 | 0.005 | 2.434 | 0.702 | 1.140 | 1.252 | 0.253 | 0.005 | 0.005 | 0.125 | 769 |
| traffic | 21/1/16 9:30 AM | 17.978 | 3.401 | 2.088 | 0.287 | 0.085 | 0.593 | 2.017 | 0.005 | 0.005 | 0.005 | 0.539 | 0.308 | 0.005 | 0.006 | 0.055 | 0.005 | 0.006 | 0.076 | 0.125 | 0.005 | 0.005 | 0.125 | 444 |
| | 26/1/16 10:45 AM | 24.041 | 7.546 | 4.441 | 0.053 | 1.393 | 2.493 | 4.795 | 0.005 | 0.205 | 0.103 | 1.190 | 0.634 | 0.134 | 0.091 | 0.105 | 0.233 | 0.177 | 0.212 | 0.125 | 0.005 | 0.005 | 0.125 | 101 |
| | 28/1/16 10:13 AM | 5.574 | 1.710 | 1.127 | 0.125 | 0.369 | 0.446 | 0.895 | 0.071 | 0.103 | 0.089 | 0.296 | 0.201 | 0.119 | 0.194 | 0.093 | 0.116 | 0.096 | 0.148 | 0.125 | 0.099 | 0.090 | 0.125 | 106 |
| | 2/2/16 8:45 AM | 29.496 | 9.643 | 5.946 | 0.175 | 1.974 | 2.209 | 5.136 | 0.068 | 0.374 | 0.245 | 1.353 | 0.791 | 0.180 | 0.334 | 0.312 | 0.177 | 0.024 | 0.216 | 0.125 | 0.086 | 0.069 | 0.125 | 80 |
| | 4/2/16 12:30 PM | 1.487 | 0.490 | 0.270 | 0.131 | 0.113 | 0.172 | 0.341 | 0.075 | 0.065 | 0.054 | 0.128 | 0.082 | 0.082 | 0.127 | 0.065 | 0.091 | 0.080 | 0.126 | 0.125 | 0.088 | 0.076 | 0.125 | 101 |
| | 9/2/16 9:15 AM | 22.421 | 6.334 | 3.563 | 0.168 | 1.058 | 1.071 | 3.330 | 0.139 | 0.092 | 0.005 | 0.849 | 0.505 | 0.345 | 0.238 | 0.389 | 0.055 | 0.130 | 0.345 | 0.409 | 0.430 | 0.641 | 0.324 | 64 |
| | 10/2/16 12:0 PM | 4.764 | 1.306 | 0.728 | 0.077 | 0.233 | 0.170 | 0.614 | 0.050 | 0.055 | 0.005 | 0.141 | 0.092 | 0.111 | 0.050 | 0.100 | 0.005 | 0.027 | 0.118 | 0.125 | 0.157 | 0.232 | 0.125 | 180 |
| G03 | 19/1/16 10:35 AM | | | | | | | | | | No PAH measurement | | | | | | | | | | | | | 639 |
| industry | 22/1/16 11:5 AM | 82.380 | 17.702 | 12.447 | 0.112 | 2.144 | 5.527 | 15.773 | 0.005 | 0.593 | 0.005 | 3.576 | 2.378 | 0.492 | 0.716 | 0.715 | 0.373 | 0.516 | 0.497 | 0.646 | 0.437 | 0.410 | 0.369 | 323 |
| | 26/1/16 8:30 AM | 25.784 | 6.766 | 4.366 | 0.005 | 1.699 | 2.954 | 5.758 | 0.039 | 0.349 | 0.083 | 1.351 | 0.716 | 0.132 | 0.219 | 0.206 | 0.209 | 0.194 | 0.220 | 0.125 | 0.088 | 0.005 | 0.125 | 85 |
| | 28/1/16 8:0 AM | 41.742 | 11.681 | 7.379 | 0.213 | 2.061 | 3.291 | 5.632 | 0.210 | 0.475 | 0.332 | 1.836 | 1.265 | 0.453 | 0.749 | 0.826 | 0.523 | 0.534 | 0.510 | 0.292 | 0.216 | 0.177 | 0.125 | 97 |
| | 2/2/16 8:0 AM | 42.695 | 11.599 | 7.393 | 0.218 | 1.897 | 3.134 | 8.855 | 0.158 | 0.517 | 0.360 | 2.393 | 1.655 | 0.475 | 0.744 | 0.737 | 0.505 | 0.507 | 0.531 | 0.264 | 0.184 | 0.140 | 0.125 | 98 |
| | 4/2/16 8:0 AM | 40.190 | 11.601 | 7.391 | 0.218 | 2.030 | 3.389 | 9.279 | 0.196 | 0.533 | 0.402 | 2.194 | 1.412 | 0.273 | 0.321 | 0.535 | 0.382 | 0.389 | 0.369 | 0.125 | 0.150 | 0.123 | 0.125 | 115 |
| | 9/2/16 9:35 AM | 45.793 | 11.529 | 7.203 | 0.339 | 2.238 | 3.509 | 10.278 | 0.163 | 1.368 | 0.271 | 3.768 | 2.869 | 1.181 | 1.523 | 1.523 | 0.693 | 0.827 | 0.864 | 0.661 | 0.551 | 0.707 | 0.454 | 118 |
| | 11/2/16 8:10 AM | 45.913 | 11.067 | 7.025 | 0.874 | 0.852 | 0.837 | 4.224 | 0.172 | 0.254 | 0.137 | 1.652 | 1.158 | 0.752 | 0.982 | 1.114 | 0.555 | 0.622 | 0.755 | 0.403 | 0.372 | 0.505 | 0.274 | 185 |
| G04 | 19/1/16 8:45 AM | 25.072 | 5.042 | 3.473 | 1.298 | 0.149 | 0.590 | 2.250 | 0.005 | 0.053 | 0.005 | 1.072 | 0.804 | 0.292 | 0.420 | 0.387 | 0.169 | 0.266 | 0.341 | 0.125 | 0.096 | 0.006 | 0.125 | 620 |
| industry | 21/1/16 8:40 AM | 21.900 | 3.922 | 2.707 | 1.152 | 0.149 | 0.608 | 2.218 | 0.005 | 0.021 | 0.005 | 0.620 | 0.489 | 0.114 | 0.120 | 0.131 | 0.006 | 0.106 | 0.139 | 0.125 | 0.005 | 0.005 | 0.125 | 285 |
| | 26/1/16 7:55 AM | 8.390 | 1.750 | 0.973 | 0.005 | 0.069 | 0.283 | 1.017 | 0.005 | 0.005 | 0.005 | 0.105 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.006 | 0.125 | 0.005 | 0.005 | 0.125 | 103 |
| | 28/1/16 8:5 AM | 4.370 | 1.275 | 0.876 | 0.233 | 0.184 | 0.283 | 0.715 | 0.080 | 0.111 | 0.083 | 0.295 | 0.208 | 0.168 | 0.238 | 0.181 | 0.183 | 0.145 | 0.190 | 0.125 | 0.152 | 0.105 | 0.125 | 107 |
| | 2/2/16 8:0 AM | 0.736 | 0.232 | 0.182 | 0.108 | 0.164 | 0.128 | 0.298 | 0.084 | 0.077 | 0.060 | 0.169 | 0.109 | 0.119 | 0.174 | 0.098 | 0.122 | 0.107 | 0.146 | 0.125 | 0.118 | 0.098 | 0.125 | 47 |
| | 4/2/16 8:10 AM | 0.600 | 0.173 | 0.122 | 0.087 | 0.121 | 0.104 | 0.204 | 0.070 | 0.044 | 0.055 | 0.132 | 0.080 | 0.098 | 0.144 | 0.073 | 0.101 | 0.087 | 0.126 | 0.125 | 0.104 | 0.087 | 0.125 | 47 |
| | 9/2/16 10:30 AM | 0.276 | 0.137 | 0.050 | 0.091 | 0.120 | 0.005 | 0.165 | 0.124 | 0.005 | 0.005 | 0.127 | 0.078 | 0.278 | 0.172 | 0.322 | 0.177 | 0.204 | 0.324 | 0.332 | 0.408 | 0.518 | 0.375 | 84 |
| | 11/2/16 8:40 AM | 5.397 | 1.321 | 0.857 | 0.084 | 0.170 | 0.166 | 0.610 | 0.142 | 0.059 | 0.005 | 0.180 | 0.128 | 0.297 | 0.171 | 0.305 | 0.172 | 0.288 | 0.332 | 0.662 | 0.635 | 0.827 | 0.378 | 170 |
| G05 | 19/1/16 10:59 AM | | | | | | | | | | No PAH measurement | | | | | | | | | | | | | 769 |
| backgrnd | 21/1/16 10:50 AM | | | | | | | | | | | | | | | | | | | | | | | 319 |
| | 26/1/16 10:20 AM | 0.026 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.063 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.005 | 0.125 | 0.005 | 0.005 | 0.125 | 119 |
| | 28/1/16 9:50 AM | 27.875 | 7.138 | 4.644 | 0.187 | 1.324 | 2.075 | 6.799 | 0.086 | 0.438 | 0.201 | 1.946 | 1.203 | 0.376 | 0.893 | 0.635 | 0.285 | 0.396 | 0.380 | 0.125 | 0.160 | 0.115 | 0.125 | 108 |
| | 2/2/16 9:32 AM | 20.717 | 5.943 | 3.707 | 0.261 | 0.873 | 1.510 | 4.267 | 0.086 | 0.405 | 0.191 | 1.344 | 0.890 | 0.244 | 0.513 | 0.354 | 0.235 | 0.242 | 0.274 | 0.125 | 0.150 | 0.113 | 0.125 | 93 |
| | 4/2/16 10:33 AM | 3.534 | 0.920 | 0.618 | 0.108 | 0.177 | 0.269 | 0.712 | 0.068 | 0.069 | 0.075 | 0.358 | 0.213 | 0.137 | 0.214 | 0.144 | 0.126 | 0.122 | 0.156 | 0.125 | 0.111 | 0.094 | 0.125 | 105 |
| | 9/2/16 9:55 AM | 11.244 | 2.495 | 1.574 | 0.094 | 0.407 | 0.455 | 2.096 | 0.103 | 0.052 | 0.005 | 0.660 | 0.392 | 0.397 | 0.350 | 0.454 | 0.222 | 0.277 | 0.385 | 0.468 | 0.421 | 0.560 | 0.323 | |
| | 11/2/16 10:20 AM | 0.101 | 0.061 | 0.022 | 0.083 | 0.065 | 0.005 | 0.061 | 0.092 | 0.005 | 0.005 | 0.057 | 0.021 | 0.209 | 0.069 | 0.203 | 0.065 | 0.111 | 0.223 | 0.366 | 0.353 | 0.475 | 0.310 | 224 |

*Table 20 PAHs and levoglucosan (LEV) levels in Menen during the winter (Jan-Feb) campaign, given in ng/m$^3$. Gradual color scale of which green indicates the lowest and red the largest value. The color scale was applied to each PAH individually but over all areas and all seasons. Each line is an individual sample for which the column 'start-PAH' gives the start time of the 48h-sampling*

| ID | start-PAK | NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP | LEV |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| M11 | 21/1/16 2:23 PM | 29.662 | 7.564 | 4.451 | 0.169 | 0.692 | 1.327 | 3.977 | 0.123 | 0.229 | 0.060 | 1.074 | 0.725 | 0.251 | 0.219 | 0.118 | 0.005 | 0.005 | 0.117 | 0.125 | 0.247 | 0.401 | 0.125 | 500 |
| industry | 26/1/16 12:43 PM | 9.111 | 2.116 | 1.351 | 0.079 | 0.318 | 0.432 | 1.607 | 0.083 | 0.022 | 0.005 | 0.450 | 0.239 | 0.154 | 0.033 | 0.071 | 0.005 | 0.005 | 0.113 | 0.125 | 0.239 | 0.388 | 0.125 | 85 |
| | 28/1/16 12:45 PM | 9.707 | 1.491 | 1.381 | 0.070 | 0.238 | 0.356 | 1.383 | 0.084 | 0.021 | 0.005 | 0.426 | 0.237 | 0.145 | 0.042 | 0.060 | 0.005 | 0.005 | 0.113 | 0.125 | 0.240 | 0.389 | 0.125 | 99 |
| | 2/2/16 12:40 PM | 7.589 | 1.846 | 1.127 | 0.073 | 0.209 | 0.185 | 0.773 | 0.087 | 0.036 | 0.005 | 0.139 | 0.075 | 0.142 | 0.005 | 0.059 | 0.005 | 0.005 | 0.113 | 0.125 | 0.238 | 0.387 | 0.125 | 203 |
| | 4/2/16 12:52 PM | 8.700 | 2.248 | 1.376 | 0.086 | 0.314 | 0.338 | 1.408 | 0.086 | 0.006 | 0.005 | 0.331 | 0.185 | 0.139 | 0.005 | 0.059 | 0.005 | 0.005 | 0.113 | 0.125 | 0.240 | 0.389 | 0.125 | 160 |
| | 9/2/16 12:37 PM | 12.366 | 2.910 | 1.879 | 0.141 | 0.340 | 0.410 | 1.428 | 0.102 | 0.027 | 0.005 | 0.409 | 0.278 | 0.162 | 0.026 | 0.071 | 0.005 | 0.005 | 0.113 | 0.125 | 0.238 | 0.387 | 0.125 | 368 |
| | 11/2/16 12:44 PM | 23.755 | 5.686 | 3.663 | 0.336 | 0.602 | 1.006 | 3.119 | 0.123 | 0.198 | 0.028 | 0.958 | 0.718 | 0.314 | 0.263 | 0.114 | 0.005 | 0.005 | 0.113 | 0.125 | 0.239 | 0.387 | 0.125 | 614 |
| | 16/2/16 12:25 PM | 22.124 | 5.549 | 3.584 | 0.150 | 0.572 | 0.933 | 2.848 | 0.104 | 0.262 | 0.005 | 0.827 | 0.609 | 0.312 | 0.268 | 0.104 | 0.005 | 0.005 | 0.112 | 0.125 | 0.237 | 0.385 | 0.125 | 624 |
| M02 | 21/1/16 1:0 PM | 54.945 | 16.026 | 10.230 | 0.376 | 1.203 | 3.055 | 8.758 | 0.177 | 0.800 | 0.326 | 2.425 | 1.886 | 0.475 | 0.738 | 0.496 | 0.079 | 0.093 | 0.211 | 0.125 | 0.195 | 0.317 | 0.125 | 566 |
| industry | 26/1/16 7:0 AM | 18.648 | 3.983 | 3.080 | 0.186 | 0.564 | 0.993 | 2.846 | 0.085 | 0.149 | 0.051 | 0.851 | 0.579 | 0.174 | 0.169 | 0.190 | 0.005 | 0.030 | 0.146 | 0.125 | 0.194 | 0.316 | 0.125 | 125 |
| | 28/1/16 7:30 AM | 18.969 | 4.734 | 3.181 | 3.159 | 0.444 | 0.859 | 2.652 | 0.066 | 0.200 | 0.089 | 0.757 | 0.492 | 0.205 | 0.295 | 0.205 | 0.005 | 0.053 | 0.138 | 0.125 | 0.192 | 0.319 | 0.125 | 140 |
| | 2/2/16 6:0 PM | 20.097 | 4.690 | 2.991 | 0.121 | 0.518 | 0.871 | 2.555 | 0.084 | 0.190 | 0.026 | 0.748 | 0.531 | 0.207 | 0.202 | 0.212 | 0.005 | 0.029 | 0.132 | 0.125 | 0.192 | 0.312 | 0.125 | 249 |
| | 4/2/16 6:45 PM | 22.241 | 5.947 | 3.766 | 0.171 | 0.558 | 1.221 | 3.667 | 0.094 | 0.278 | 0.086 | 0.985 | 0.681 | 0.192 | 0.255 | 0.222 | 0.005 | 0.006 | 0.135 | 0.125 | 0.196 | 0.319 | 0.125 | 250 |
| | 9/2/16 7:15 AM | 25.415 | 6.042 | 3.984 | 0.140 | 0.604 | 1.045 | 3.250 | 0.175 | 0.388 | 0.087 | 0.830 | 0.639 | 0.253 | 0.320 | 0.251 | 0.005 | 0.046 | 0.153 | 0.125 | 0.195 | 0.317 | 0.125 | 312 |
| | 11/2/16 7:15 AM | 40.555 | 12.773 | 8.572 | 1.547 | 1.118 | 2.136 | 6.093 | 0.169 | 0.587 | 0.152 | 1.561 | 1.222 | 0.437 | 0.617 | 0.348 | 0.029 | 0.031 | 0.164 | 0.125 | 0.192 | 0.312 | 0.125 | 606 |
| | 16/2/16 8:30 AM | 44.180 | 14.774 | 9.809 | 0.568 | 1.478 | 2.541 | 7.344 | 0.153 | 0.815 | 0.222 | 2.236 | 1.813 | 0.646 | 0.846 | 0.505 | 0.122 | 0.071 | 0.228 | 0.125 | 0.180 | 0.293 | 0.125 | 766 |
| M03 | 21/1/16 10:50 AM | 43.633 | 12.095 | 8.049 | 0.259 | 0.868 | 2.182 | 5.969 | 0.120 | 0.322 | 0.192 | 1.424 | 1.008 | 0.359 | 0.466 | 0.332 | 0.063 | 0.044 | 0.137 | 0.125 | 0.198 | 0.321 | 0.125 | 536 |
| backgrnd | 26/1/16 6:50 AM | 12.919 | 3.032 | 2.439 | 0.092 | 0.498 | 0.824 | 2.089 | 0.072 | 0.208 | 0.006 | 0.532 | 0.326 | 0.151 | 0.097 | 0.118 | 0.005 | 0.005 | 0.123 | 0.125 | 0.200 | 0.325 | 0.125 | 90 |
| | 28/1/16 7:10 AM | 16.469 | 1.843 | 2.958 | 0.102 | 0.533 | 0.882 | 2.300 | 0.080 | 0.148 | 0.027 | 0.556 | 0.377 | 0.157 | 0.092 | 0.129 | 0.005 | 0.005 | 0.120 | 0.125 | 0.194 | 0.315 | 0.125 | 132 |
| | 2/2/16 6:45 AM | 15.097 | 5.177 | 3.714 | 0.123 | 0.473 | 0.608 | 1.632 | 0.081 | 0.111 | 0.005 | 0.569 | 0.334 | 0.172 | 0.101 | 0.124 | 0.005 | 0.005 | 0.117 | 0.125 | 0.199 | 0.322 | 0.125 | 94 |
| | 4/2/16 7:25 AM | 16.711 | 4.852 | 3.176 | 0.098 | 0.386 | 0.877 | 2.299 | 0.070 | 0.104 | 0.006 | 0.639 | 0.382 | 0.148 | 0.137 | 0.115 | 0.005 | 0.005 | 0.116 | 0.125 | 0.196 | 0.319 | 0.125 | 149 |
| | 9/2/16 6:50 AM | 19.619 | 5.967 | 4.131 | 0.166 | 0.495 | 0.933 | 2.032 | 0.070 | 0.144 | 0.005 | 0.553 | 0.377 | 0.174 | 0.172 | 0.158 | 0.005 | 0.005 | 0.112 | 0.125 | 0.194 | 0.315 | 0.125 | 159 |
| | 11/2/16 8:40 AM | 35.279 | 9.595 | 6.443 | 0.548 | 0.716 | 1.498 | 3.561 | 0.099 | 0.243 | 0.073 | 0.909 | 0.697 | 0.388 | 0.454 | 0.322 | 0.058 | 0.005 | 0.146 | 0.125 | 0.201 | 0.326 | 0.125 | 614 |
| | 16/2/16 6:45 AM | 38.744 | 11.774 | 7.795 | 0.165 | 0.619 | 1.779 | 4.089 | 0.098 | 0.346 | 0.079 | 1.217 | 0.915 | 0.508 | 0.651 | 0.397 | 0.075 | 0.006 | 0.157 | 0.125 | 0.200 | 0.325 | 0.125 | 686 |
| M04 | 21/1/16 11:50 AM | 38.284 | 9.681 | 6.541 | 0.164 | 0.711 | 1.924 | 5.650 | 0.116 | 0.339 | 0.156 | 1.618 | 1.152 | 0.399 | 0.491 | 0.300 | 0.035 | 0.005 | 0.148 | 0.125 | 0.196 | 0.318 | 0.125 | 655 |
| backgrnd | 26/1/16 8:55 AM | 10.753 | 3.172 | 1.994 | 0.086 | 0.251 | 0.465 | 1.593 | 0.070 | 0.093 | 0.005 | 0.428 | 0.280 | 0.137 | 0.082 | 0.093 | 0.005 | 0.005 | 0.093 | 0.125 | 0.196 | 0.318 | 0.125 | 116 |
| | 28/1/16 9:0 AM | 12.285 | 3.127 | 1.905 | 0.064 | 0.251 | 0.418 | 1.391 | 0.067 | 0.058 | 0.005 | 0.389 | 0.259 | 0.123 | 0.064 | 0.087 | 0.005 | 0.005 | 0.092 | 0.125 | 0.195 | 0.316 | 0.125 | 165 |
| | 2/2/16 9:15 AM | 11.648 | 3.367 | 2.117 | 0.086 | 0.320 | 0.384 | 1.177 | 0.074 | 0.098 | 0.005 | 0.329 | 0.224 | 0.138 | 0.021 | 0.073 | 0.005 | 0.005 | 0.093 | 0.125 | 0.196 | 0.318 | 0.125 | 113 |
| | 4/2/16 9:20 AM | 12.629 | 3.481 | 2.110 | 0.102 | 0.294 | 0.586 | 1.841 | 0.076 | 0.068 | 0.005 | 0.503 | 0.362 | 0.146 | 0.069 | 0.081 | 0.005 | 0.005 | 0.093 | 0.125 | 0.196 | 0.318 | 0.125 | 183 |
| | 9/2/16 9:0 AM | 15.505 | 4.134 | 2.679 | 0.120 | 0.380 | 0.545 | 1.661 | 0.072 | 0.066 | 0.005 | 0.471 | 0.312 | 0.156 | 0.076 | 0.096 | 0.005 | 0.005 | 0.093 | 0.125 | 0.196 | 0.318 | 0.125 | 217 |
| | 11/2/16 9:0 AM | 27.965 | 7.452 | 4.968 | 0.793 | 0.648 | 1.184 | 3.291 | 0.104 | 0.298 | 0.063 | 0.919 | 0.726 | 0.319 | 0.314 | 0.152 | 0.005 | 0.005 | 0.093 | 0.125 | 0.196 | 0.318 | 0.125 | 635 |
| | 16/2/16 9:15 AM | 29.975 | 8.931 | 5.929 | 0.401 | 0.682 | 1.437 | 3.852 | 0.122 | 0.397 | 0.097 | 1.144 | 0.878 | 0.393 | 0.433 | 0.159 | 0.005 | 0.005 | 0.093 | 0.125 | 0.196 | 0.318 | 0.125 | 698 |
| M05 | 21/1/16 8:50 AM | 29.901 | 7.975 | 5.154 | 0.214 | 0.663 | 1.285 | 3.918 | 0.097 | 0.212 | 0.111 | 1.061 | 0.770 | 0.290 | 0.306 | 0.142 | 0.005 | 0.005 | 0.095 | 0.125 | 0.201 | 0.327 | 0.125 | 535 |
| traffic | 26/1/16 10:30 AM | 9.098 | 4.764 | 2.994 | 0.105 | 0.462 | 0.446 | 1.305 | 0.075 | 0.005 | 0.005 | 0.324 | 0.183 | 0.130 | 0.020 | 0.063 | 0.005 | 0.005 | 0.098 | 0.125 | 0.208 | 0.338 | 0.125 | 79 |
| | 28/1/16 9:20 AM | 1.438 | 0.365 | 0.208 | 0.058 | 0.070 | 0.005 | 0.092 | 0.078 | 0.005 | 0.005 | 0.005 | 0.005 | 0.106 | 0.005 | 0.044 | 0.005 | 0.005 | 0.094 | 0.125 | 0.199 | 0.322 | 0.125 | 105 |
| | 2/2/16 9:10 AM | 11.108 | 2.755 | 1.705 | 0.086 | 0.313 | 0.386 | 1.244 | 0.079 | 0.006 | 0.005 | 0.347 | 0.228 | 0.140 | 0.021 | 0.067 | 0.005 | 0.005 | 0.097 | 0.125 | 0.204 | 0.332 | 0.125 | 140 |
| | 4/2/16 8:50 AM | 7.402 | 2.309 | 1.488 | 0.126 | 0.184 | 0.278 | 0.926 | 0.076 | 0.031 | 0.005 | 0.197 | 0.101 | 0.133 | 0.022 | 0.074 | 0.005 | 0.005 | 0.093 | 0.125 | 0.196 | 0.318 | 0.125 | 158 |
| | 9/2/16 9:15 AM | 16.274 | 3.823 | 2.449 | 0.143 | 0.343 | 0.496 | 1.649 | 0.090 | 0.035 | 0.005 | 0.401 | 0.278 | 0.176 | 0.097 | 0.099 | 0.005 | 0.005 | 0.097 | 0.125 | 0.206 | 0.334 | 0.125 | 246 |
| | 11/2/16 8:40 AM | 4.937 | 1.522 | 0.945 | 0.137 | 0.118 | 0.005 | 0.404 | 0.102 | 0.036 | 0.005 | 0.083 | 0.044 | 0.116 | 0.005 | 0.043 | 0.005 | 0.005 | 0.094 | 0.125 | 0.198 | 0.322 | 0.125 | 471 |
| | 16/2/16 9:10 AM | 27.817 | 8.036 | 5.063 | 0.179 | 1.285 | 1.637 | 4.490 | 0.116 | 0.327 | 0.089 | 2.352 | 1.480 | 1.053 | 1.342 | 0.243 | 0.006 | 0.005 | 0.098 | 0.125 | 0.207 | 0.337 | 0.125 | 608 |
| M06 | 21/1/16 8:30 AM | 7.331 | 1.621 | 1.067 | 0.885 | 0.160 | 0.172 | 0.792 | 0.096 | 0.109 | 0.005 | 0.251 | 0.184 | 0.197 | 0.089 | 0.072 | 0.005 | 0.005 | 0.098 | 0.125 | 0.208 | 0.338 | 0.125 | 746 |
| traffic | 26/1/16 8:0 AM | Technical problem PAH measurment | | | | | | | | | | | | | | | | | | | | | | 19 |
| | 28/1/16 5:0 PM | | | | | | | | | | | | | | | | | | | | | | | 108 |
| | 2/2/16 7:0 AM | | | | | | | | | | | | | | | | | | | | | | | 156 |
| | 4/2/16 2:30 PM | | | | | | | | | | | | | | | | | | | | | | | 171 |
| | 9/2/16 6:30 AM | | | | | | | | | | | | | | | | | | | | | | | 200 |
| | 11/2/16 4:30 PM | | | | | | | | | | | | | | | | | | | | | | | 599 |
| | 16/2/16 7:30 AM | | | | | | | | | | | | | | | | | | | | | | | 660 |

*Table 21 PAHs and levoglucosan (LEV) levels in Houtem during the winter (Jan-Feb) campaign given in ng/m$^3$. Gradual color scale of which green indicates the lowest and red the largest value. The color scale was applied to each PAH individually but over all areas and all seasons. Each line is an individual sample for which the column 'start-PAH' gives the start time of the 48h-sampling*

| ID | start-PAK | NAP | 2MeNAP | 1MeNAP | ACY | ACE | FLU | PHE | 9MePHE | ANT | 2MeANT | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC | BghiP | LEV |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| H backgrnd | 21/1/16 6:50 AM | 58.866 | 13.426 | 9.335 | 0.264 | 1.443 | 3.607 | 8.763 | 0.134 | 0.379 | 0.219 | 2.647 | 1.733 | 0.574 | 0.964 | 0.997 | 0.268 | 0.350 | 0.440 | 0.100 | 0.256 | 0.361 | 0.100 | 783 |
| | 26/1/16 8:30 AM | 9.787 | 2.467 | 1.547 | 0.066 | 0.330 | 0.618 | 1.784 | 0.080 | 0.174 | 0.005 | 0.433 | 0.279 | 0.191 | 0.079 | 0.227 | 0.005 | 0.052 | 0.186 | 0.100 | 0.236 | 0.372 | 0.100 | 54 |
| | 28/1/16 7:8 AM | 10.742 | 1.603 | 1.511 | 0.065 | 0.295 | 0.457 | 1.324 | 0.075 | 0.174 | 0.005 | 0.357 | 0.258 | 0.161 | 0.129 | 0.205 | 0.005 | 0.048 | 0.171 | 0.100 | 0.215 | 0.340 | 0.100 | 65 |
| | 2/2/16 6:45 AM | 7.294 | 1.836 | 1.064 | 0.074 | 0.381 | 0.285 | 0.923 | 0.082 | 0.120 | 0.005 | 0.320 | 0.193 | 0.190 | 0.063 | 0.195 | 0.005 | 0.042 | 0.171 | 0.100 | 0.227 | 0.353 | 0.100 | 27 |
| | 4/2/16 7:12 AM | 26.631 | 5.495 | 3.690 | 0.367 | 0.562 | 1.522 | 5.109 | 0.121 | 0.564 | 0.116 | 1.565 | 1.169 | 0.438 | 0.418 | 0.474 | 0.086 | 0.128 | 0.304 | 0.100 | 0.215 | 0.328 | 0.100 | 86 |
| | 9/2/16 7:0 AM | 33.558 | 6.989 | 4.347 | 0.121 | 1.049 | 2.151 | 4.483 | 0.090 | 0.256 | 0.070 | 1.499 | 1.047 | 0.522 | 0.539 | 0.603 | 0.130 | 0.166 | 0.311 | 0.100 | 0.227 | 0.347 | 0.100 | 65 |
| | 11/2/16 7:11 AM | 35.153 | 8.457 | 5.456 | 0.218 | 1.067 | 1.875 | 4.271 | 0.088 | 0.221 | 0.059 | 1.299 | 0.772 | 0.356 | 0.467 | 0.584 | 0.129 | 0.153 | 0.306 | 0.100 | 0.217 | 0.340 | 0.100 | 279 |
| | 16/2/16 6:48 AM | 40.672 | 9.336 | 6.289 | 0.215 | 1.207 | 2.252 | 5.835 | 0.120 | 0.378 | 0.104 | 1.748 | 1.210 | 0.487 | 0.682 | 0.635 | 0.156 | 0.163 | 0.318 | 0.100 | 0.213 | 0.345 | 0.100 | 158 |

### 3.4.9. POSSIBLE INFLUENCE OF TRAFFIC WORKS, STREET FESTIVALS IN MENEN

The Ieperstraat in Menen was closed during one day for a street festival on the 5th of September 2015. Measurements were ongoing in the neighborhood of the Ieperstraat. The closest monitoring station M05 (traffic station) recorded 48h measurements on the 1st, 3th and 8th of September. Measurements at M05, were for all PAHs on those days clearly lower than for the other days. It might be that traffic was influenced due to presence of road signs, but it seems unlikely that the influence could be of a that long duration.

During the period of the 31th of August 2015 and the first of October 2015 the Hogeweg was blocked because of traffic works on the roundabout Hogeweg, Wervikstraat and Wilgenlaan. A deviation was foreseen. This may have had an influence on measurements in the neighborhood. Since the deviation covered the entire summer-fall measuring campaign, the measurement variation over the specified time period could not be caused by this deviation. The impact on the seasonal variation, is difficult to estimate.

### 3.4.10. INFLUENCE OF INDUSTRY WITH KNOWN PAH EMISSIONS

During the current study, emissions from some of the companies in the neighborhood of the measuring units were measured. The dossiers were checked for PAH emissions which might have influenced PAH levels observed on the monitoring stations of the current study.

**Menen**

In three companies A, B and C located in Menen (with a.o. known would burning emissions), the PHE emissions were pronounced (Table 22). The average PHE immission levels measured in the current summer-fall campaign was, for stations classified as industry and background, respectively 4.1 (stdev 3.3) ng/m$^3$ and 5.8 (st.dev. 2.6) ng/m$^3$. In winter the levels were 3.5 (st.dev. 2.2) ng/m$^3$ for the industry stations and 2.8 (stdev 1.5) ng/m$^3$ on the background locations in Menen. There was no significant difference in PHE concentrations between the industry and background stations. In other words, the impact of the companies was not observed. PHE has indeed also other ubiquitous sources, such as emissions from diesel, gasoline and wood burning (Boström et al., 2002).

Company C emitted relative larger PAH concentrations than the other companies. The company mainly emitted NAP (55% of total PAHs), ACY (20%) aside from PHE (7%). Two of these PAHs were relatively higher present in all profiles of Menen measured in the current study: NAP (Figure 9, PAH number 1 in x-axis) and PHE (PAH number 7 in x-axis) but not ACY (PAH number 4 in X-axis). It was not different from the profiles in Genk, indicating that the impact of the company observed on our measuring stations was not significant when considering these profiles. Nevertheless PHE and/or 9MePHE appeared clearly in the profile of M01, M02, M03, M04, M05 and M06. The median concentration of NAP was in the summer-fall period higher in Menen compared to Genk: 20.25 vs. 12.99 ng/m$^3$ (Table 7, in winter this was not observed, but in that period NAP is influenced by wood burning). PHE concentrations were similar in both regions, and ACY was higher in Genk during the summer-fall measurements (Table 7).

*Table 22 PAHs measured at three industrial sites in Menen, given in µg/Nm$^3$. Date of the measurements: Company A: February'15, Company B: January'15, Company C: March/April'15*

| Company | NAP | 2MeNAP | 1MeNAP* | ACY | ACE | FLU | PHE | 9MePHE* | ANT | 2MeANT* | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC* | BghiP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **A** | < | < |  | < | < | < | 1.7 |  | < |  | < | < | < | < | < | < | < | < | < | < |  | < |
| **B** | 6.4 | < |  | < | < | 0.9 | 6.8 |  | < |  | 0.7 | < | < | < | < | < | < | < | < | < |  | < |
| **C**** | 84000 | 1560 |  | 30000 | 63 | 1430 | 11000 |  | 1810 |  | 6300 | 7100 | 650 | 650 | 1040 | 350 | 1690 | 2900 | 1300 | 56 |  | 970 |

**: Not measured, <: Below specified value, **: results were out of range of the linear limit of the used method*

### Genk

At the industrial site of Company D (burning of off-gasses) in Genk, only PHE was emitted, at levels comparable to company A of Menen mentioned above. The steel company E showed relatively larger values for NAP, ACY, FLU and PHE (NB: it was checked if the periods of inactivity were visible in the low molecular PAH profiles of Genk, but this was not the case). Similar PAHs and similar concentrations were emitted as company B in Menen. Factor analysis for the Genk summer-fall data identified a factor enriched with ACY and ACE (equal contribution), typical for steel and iron industry (see 3.5.2). ACY and ACE emissions were not measured in company D and E, except at the measuring campaign of March'16 (E''') (outside current study period), when ACY levels of 6.9 µg/Nm$^3$ were observed.

*Table 23: PAHs measured at an industrial site in Genk in µg/m$^3$ . Date of the measurements: Company D: March'15, Company E: August'15, Company E': November'15, Company E'': March'16, Company E''': March'16.*

| Company | NAP | 2MeNAP | 1MeNAP* | ACY | ACE | FLU | PHE | 9MePHE* | ANT | 2MeANT* | FLA | PYR | BaA | CHR | BbF | BkF | BeP | BaP | IcdP | DahA | PIC* | BghiP |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| D | < | < |  | < | < | < | 0.9 |  | < |  | < | < | < | < | < | < | < | < | < | < |  | < |
| E | 0.52 | 0.77 |  | < | < | 0.75 | 2.8 |  | < |  | < | < | < | < | < | < | < | < | < | < |  | < |
| E' | 0.93 | 2.1 |  | 0.65 | 3.1 | 10.8 | 46 |  | 4.6 |  | 1.5 | 2.4 | < | < | < | < | < | < | < | < |  | < |
| E'' | 3.7 | < |  | < | < | < | < |  | < |  | < | < | < | < | < | < | < | < | < | < |  | < |
| E''' | 40 | 2.4 |  | 6.9 | < | 2.2 | 2.4 |  | < |  | < | < | < | < | < | < | < | < | < | < |  | < |

**: Not measured, <: Below specified value. The accents for company A indicate different time points and chimneys*

## 3.5. SOURCES OF PAHS AND LEVOGLUCOSAN IN THE AIR AT THE DIFFERENT SAMPLING LOCATIONS

Different techniques exist for the source apportionment of PAHs in the environment: e.g. molecular tracers, diagnostic ratios and principal component analysis (PMF or positive matrix factorization). Often a combination of these techniques is used as the application of one technique only can be insufficient to derive strong conclusions and a combination will reinforce the results (Gao et al., 2013).

It should be noted that tools are only indicative, as even differences in sampling/analysis techniques can cause changes in the profile. Furthermore, different sources often mix up, possibly even generating (slightly) similar profiles, or due to different distance of the source, complicate the profile. Indeed, the ratio can also be altered due to ageing of the air sample due to atmospheric reactivity of some PAHs with other atmospheric species.

### 3.5.1. DIAGNOSTIC RATIOS

#### → Vehicle emissions sources: FLU/(FLU+PYR)

The ratio FLU/(FLU+PYR) is an indicative diagnostic ratio for the **type of vehicle emissions**. If this ratio is greater than 0.5, it is a measure for diesel emissions. Below 0.5, it is indicative for petrol emissions (Tobiszewski and Namiesnik, 2012). Overall, in both measuring seasons, and both regions FLU/(FLU+PYR) ratios indicated diesel emissions as the major vehicle emissions. Indeed, in summer-fall the average ratio was 0.54 and 0.69 for the traffic and background stations of Genk respectively. However, with the exception of the industry monitoring stations of Genk (i.e. ratio=0.28), indicating larger petrol emissions. In winter, the average ratio over all measuring stations in Genk was 0.60 (stdev: 0.17). In Menen, the overall average ratio during summer-fall and winter equaled 0.72 (stdev: 0.13), and 0.64 (stdev: 0.05) (NB: Only occasionally, ratios below 0.50 were observed in Genk: three times at the industry stations G03 and G04 and once at the background station G05).

#### → IcdP/(IcdP+BghiP)

The ratio IcdP/(IcdP+BghiP)[8] is indicative for petrogenic PAH sources (ratio<0.2), petroleum combustion (0.2-0.5), or gras/wood/coal combustion (>0.5) (Katsoyiannis and Breivik, 2014; Dvorská et al., 2011). Based on these ratios, the sources in Genk and Menen could be categorized as pyrogenic i.e. a mix of petroleum, biomass and coal combustion (Figure 10). This is not in accordance with the result obtained from the ANT/(ANT+PHE) ratio which categorized the samples mainly as petrogenic (Figure 11). Inconsistent results can be found based on different diagnostic ratios. The ratio IcdP/(IcdP+BghiP) is considered as stable and trustworthy (see further double ratio plots).

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[8] IcdP and BghiP values < 0.2 ng/m3 (detection limit for both PAHs) were set at half of the detection limit. In general, when the nominator or denominator in a diagnostic ratio ratio was equal to the detection limit of the PAHs, no ratios were calculated. In Menen, the majority of the values for IcdP and BghiP were below detection limit for the winter campaign which explains the few calculated ratio's.

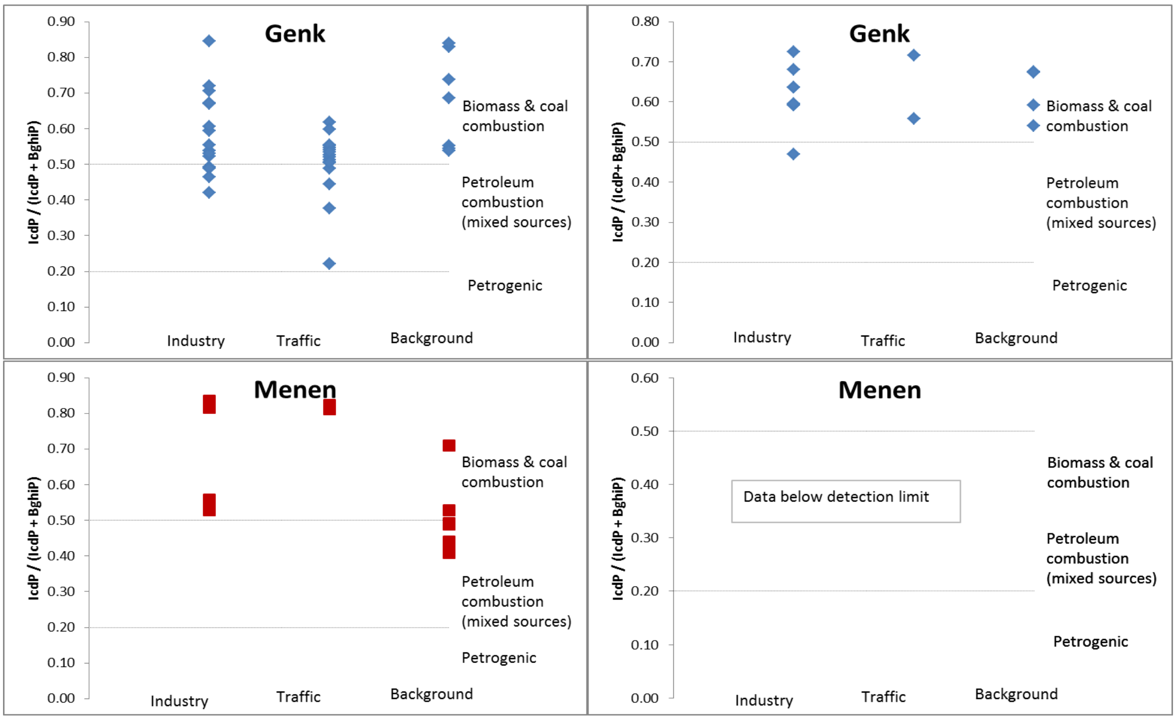


Figure 10 Ratio IcdP/(IcdP+BghiP) for all three types of monitoring stations in Genk (upper row) *Menen (lower row) during summer-fall (left) and winter (right).*

### → Double ratio plots

Double ratio plots potentially allow a more elaborate source apportionment then source apportionment based on single diagnostic ratios (Elorduy, 2016). Double ratio plots were made for ANT/(ANT+PHE) against FLA/(FLA+PYR) and BaA/(BaA+CHR) against IcdP/(IcdP+BghiP). The ratios were displayed for all sampling points of all monitoring stations of Genk and Menen. Based on the (FLA/(FLA+PYR)) ratio in Figure 11, PAH exposure in Genk during summer-fall was determined by a mix of combustion sources (petroleum, biomass and coal). In winter, the sources were dominated by biomass and coal combustion. However, most sampling locations had ANT/(ANT+PHE) ratios below 0.1, theoretically indicating petrogenic sources. In other words, based on different diagnostic ratios, inconsistent source suggestions were found. This has also been reported in scientific literature (Park et al., 2011). The efficiency of the diagnostic ratios as a source appointment instrument was previously reviewed by several researchers. The diagnostic ratios BaA/(BaA+CHR) and IcdP/(IcdP+BghiP) were recommended as comparatively robust and trustworthy (Katsoyiannis and Breivik, 2014; Dvorská et al., 2011).

Based on the double ratio plot of BaA/(BaA+CHR) against IcdP/(IcdP+BghiP) sources in Genk/Diepenbeek in summer-fall mostly categorized as pyrogenic (i.e., a mix of petroleum or biomass and coal combustion). In Genk/Diepenbeek, combustion ratios were seen on the industry and traffic locations, whereas petroleum combustion, as well as biomass and coal combustion ratios were observed on the industry and background stations (Figure 10). In summer-fall the IcdP/(IcdP+BghiP) ratios in Menen were mostly above 0.5, and for some background samples between 0.2-0.5. In winter on all locations of Genk/Diepenbeek and Menen, the double ratio plot of BaA/(BaA+CHR) against IcdP/(IcdP+BghiP), indicated combustion of biomass and coal as a primary PAH sources (Figure 12). This was confirmed by the BaA/(BaA+CHR) ratios which were almost all higher than 0.35 (combustion) in Genk/Diepenbeek (NB: ratio could not be calculated in Menen since the levels were below detection limit).

Overall based on FLA / (FLA + PYR) and IcdP / (IcdP + BghiP) ratios, samples in the summer were for both Menen and Genk/Diepenbeek identified as mainly coming from petroleum combustion, biomass and coal combustion whereas in winter the variation of the ratios decreases and was more determined by biomass and coal combustion (Figure 11; Figure 12).

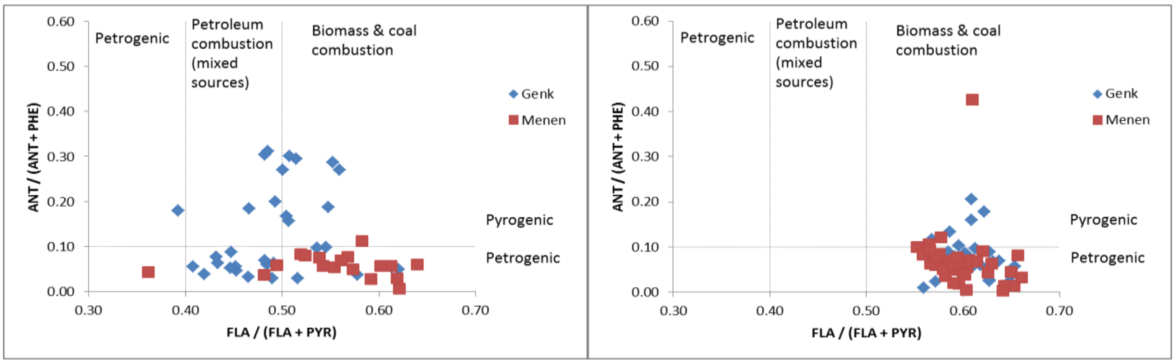


*Figure 11 Double ratio plots (for Genk/Diepenbeek and Menen) of the diagnostic ratios ANT/(ANT+PHE) versus FLA/(FLA+PYR) during summer-fall (left) and winter (right). Plot based on Elorduy2016.*

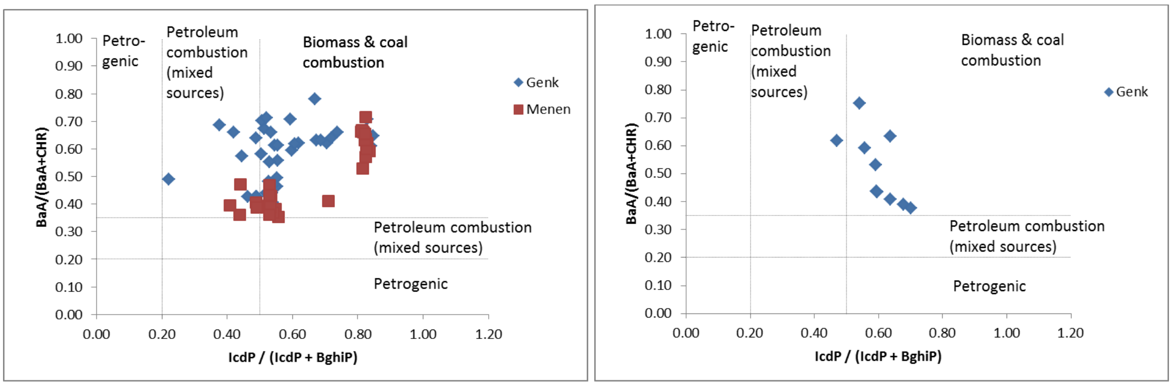


*Figure 12 Double ratio plots (for Genk/Diepenbeek and Menen) of diagnostic ratios BaA/(BaA+CHR) versus IcdP/(IcdP+BghiP) during summer-fall (left) and winter (right). Plot based on Elorduy2016.*

### 3.5.2. FACTOR ANALYSIS FOR SOURCE APPORTIONMENT

Both PCA (Principal Component Analysis with VariMax rotation) and PMF (Positive Matrix Factorisation) were done to check the rigidity of the source apportionment analysis. It was already shown by Park et al. (2011), that similar factors can be generated using both techniques. Overall, we also observed a good agreement between PCA an PMF, therefore only PCA was presented. Note: During the summer-fall (Sept'15) campaign there was no significant correlation of levoglucosan with any of the PAHs. Therefore levoglucosan was only added to the factor analysis in the winter season factor analysis. In winter, levoglucosan levels (Jan-Feb'16) were significantly

(Spearman rank correlations r=0.36-0.78, p<0.05) positive correlated with all PAHs from NAP to BaP except with 9MePHE and the 5-and 6-ring PAHs IcdP, DahP, PIC and BghiP.

**Genk**

*Summer-fall (Sept'15) campaign*

**Factor1** (43% of total variance) was loaded with ANT, 2MeANP, and high molecular weight PAHs, which pointed to vehicle emissions and traffic. Some compounds of the **second** (FLU, PHE) and **third factor** (FLA, PYR) could be explained by diesel vehicle emissions (Khalili et al., 1995). Indeed FLU, PHE, ANT, and PYR were earlier described to be related to diesel emissions (Ravindra et al., 2008) aside from 2MeANT (Westerholm & Li, 1994). **Factor 4** (7% of total variance) loaded with ACE and ACY was observed both in PMF as in the PCA analysis. These components could be indicative of the steel and iron industry which use heavy oils as fuel (Yang et al., 2002). Those low molecular weight PAHs can also originate form petrochemistry (Park et al., 2011).

*Winter (Jan-feb'16) campaign*

In the Jan-Feb campaign, the first factor was dominated by the presence of 2- tot 5-ring PAHs and levoglucosan. Therefore, **factor 1** was assigned to contain tracers for wood burning and diesel exposure. Tracers for wood burning are levoglucosan, BaA, ACE, NAP, ANT, PHE, BaP, BeP (Khalili et al., 1995). Teixeira et al. 2013 indicated that the factor loaded with ACE, FLU, PHE, ANT, PYR, BbF, BhF and BaP was representative for wood burning. Ravindra et al. (2008) reported that a high factor loadings for FLU, PHE, ANT, and PYR were related to diesel emissions. Westerholm & Li (1994) identified 2MeANT as one of the most abundant PAH compounds in the emissions from diesel fuel vehicles. In other studies it was also suggested that FLU, PYR with high factor loading of BbF and BkF indicated diesel powered vehicle emissions (Khalili et al., 1995).

The **second factor** was characterized by relatively high factor loading for IcdP, DahA, PIC and BghiP. Park et al. (2011) observed that heavy PAHs, such as IcdP, DahA, and BghiP were related to vehicular emissions. This was confirmed by Teixeira et al. (2013) and Callen et al. (2014).

The **third factor** found in the PCA analysis (high factor loading BaA and CHR) was not individually present in the PMF analysis. Park et al. (2011) identified a relatively high contribution of BaA and CHR as natural gas combustion.

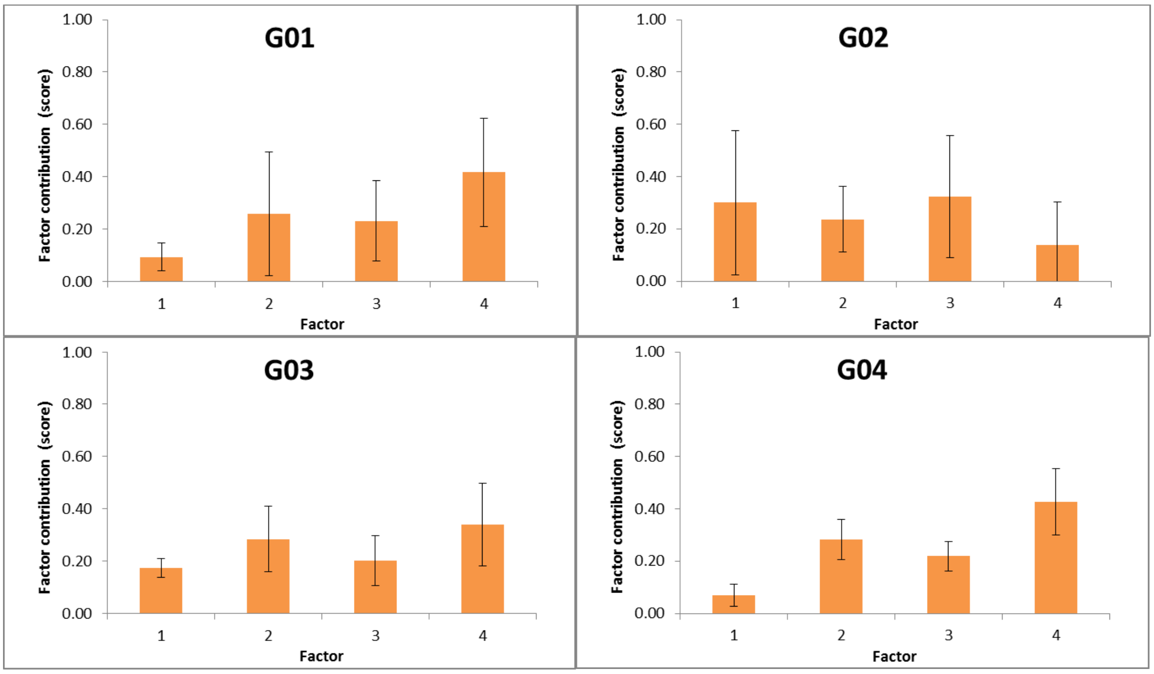
*Table 24: Factor loadings for Genk in both sampling seasons, obtained via PCA analysis (factor loadings > 0.7 are indicated in red. Those above 0.45 indicated in blue)*

| Genk | Summer-fall (Sept'15) | | | | Winter (Jan-Feb'16) | | |
|---|---|---|---|---|---|---|---|
| | Factor1 Traffic (HMW PAH) | Factor2 Traffic (LMW PAH) | Factor3 Traffic (heavy?) | Factor4 Steel or petrochemical industry | Factor1 Wood burning&traffic | Factor2 Traffic or? | Factor3 Natural gas combustion? |
| **NAP** | 0.06 | 0.80 | 0.35 | 0.13 | 0.88 | 0.18 | 0.14 |
| **2MeNAP** | 0.19 | 0.95 | 0.18 | 0.08 | 0.95 | 0.04 | 0.24 |
| **1MeNAP** | 0.20 | 0.94 | 0.16 | 0.08 | 0.95 | 0.02 | 0.26 |
| **ACY** | -0.01 | 0.06 | 0.09 | 0.89 | 0.90 | -0.16 | -0.04 |
| **ACE** | 0.03 | 0.24 | -0.24 | 0.78 | 0.92 | -0.02 | 0.31 |
| **FLU** | 0.11 | 0.91 | 0.19 | 0.11 | 0.95 | -0.03 | 0.20 |
| **PHE** | 0.13 | 0.91 | 0.32 | -0.04 | 0.96 | 0.00 | 0.21 |
| **9MePHE** | 0.17 | 0.49 | 0.77 | -0.05 | 0.43 | 0.30 | 0.69 |
| **ANT** | 0.74 | 0.40 | 0.50 | 0.02 | 0.90 | -0.15 | 0.30 |
| **2MeANT** | 0.71 | 0.60 | 0.03 | 0.09 | 0.87 | -0.01 | 0.44 |

| Genk | Summer-fall (Sept'15) | | | | Winter (Jan-Feb'16) | | |
|---|---|---|---|---|---|---|---|
| | Factor1 Traffic (HMW PAH) | Factor2 Traffic (LMW PAH) | Factor3 Traffic (heavy?) | Factor4 Steel or petrochemical industry | Factor1 Wood burning&traffic | Factor2 Traffic or? | Factor3 Natural gas combustion? |
| **FLA** | 0.18 | 0.48 | 0.83 | -0.03 | 0.96 | -0.04 | 0.22 |
| **PYR** | 0.16 | 0.32 | 0.89 | -0.01 | 0.95 | -0.01 | 0.28 |
| **BaA** | 0.80 | 0.19 | 0.50 | -0.01 | 0.37 | 0.26 | 0.87 |
| **CHR** | 0.90 | -0.04 | 0.36 | -0.01 | 0.40 | 0.13 | 0.88 |
| **BbF** | 0.90 | 0.10 | 0.38 | 0.01 | 0.90 | 0.24 | 0.31 |
| **BkF** | 0.98 | -0.04 | 0.07 | -0.03 | 0.76 | 0.34 | 0.33 |
| **BeP** | 0.87 | -0.08 | 0.44 | -0.03 | 0.85 | 0.40 | 0.09 |
| **BaP** | 0.98 | 0.02 | 0.11 | 0.01 | 0.86 | 0.40 | 0.17 |
| **IcdP** | 0.92 | 0.36 | -0.06 | 0.04 | 0.19 | 0.96 | 0.00 |
| **DahA** | 0.92 | 0.31 | -0.15 | 0.02 | -0.07 | 0.96 | 0.18 |
| **PIC** | 0.93 | 0.31 | -0.17 | 0.00 | -0.07 | 0.93 | 0.21 |
| **BghiP** | 0.88 | 0.26 | 0.19 | 0.03 | 0.00 | 0.96 | 0.07 |
| **LEV** | - | - | - | - | 0.71 | -0.17 | 0.23 |
| **Eigenvalue** | **9.51** | **5.67** | **3.52** | **1.47** | **13.31** | **4.40** | **3.11** |
| **% Variance** | **0.43** | **0.26** | **0.16** | **0.07** | **0.58** | **0.19** | **0.14** |

*HMW: high molecular weight PAH, LMW: low molecular weight*

The relative factor scores of each of the measuring units in Genk indicate which proportion each factor of the factor analysis is contributing to the PAH mixture measured on that location. It was clear that factor 4 ('steel or petrochemical industry') loaded with ACY took up an important percentage of the PAH compounds at the traffic measuring unit G01, the industry units G03 and G04 and even the background station G05 (Figure 13). In winter, the factors 1 and 2 including wood burning and traffic exposure, were the main contributors on all measuring units (Figure 14).



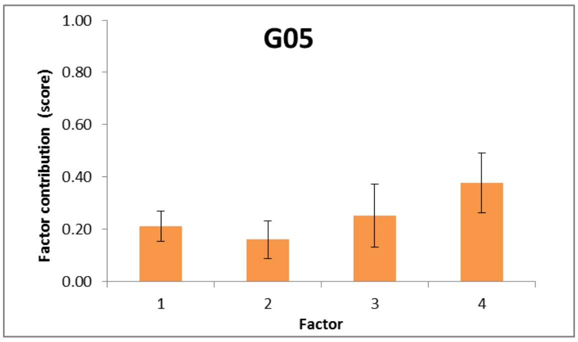


*Figure 13: Average percentage apportionments of PAHs measured in summer-fall for each of the measuring units of Genk. The relative factor scores indicate which proportion of each factor of the factor analysis is contributing to the PAH mixture measured on that location. Factor 1 = Traffic (HMW PAH), Factor 2 = Traffic (LMW PAH), Factor 3 = Traffic (heavy?), Factor 4 = Steel or petrochemical industry.*

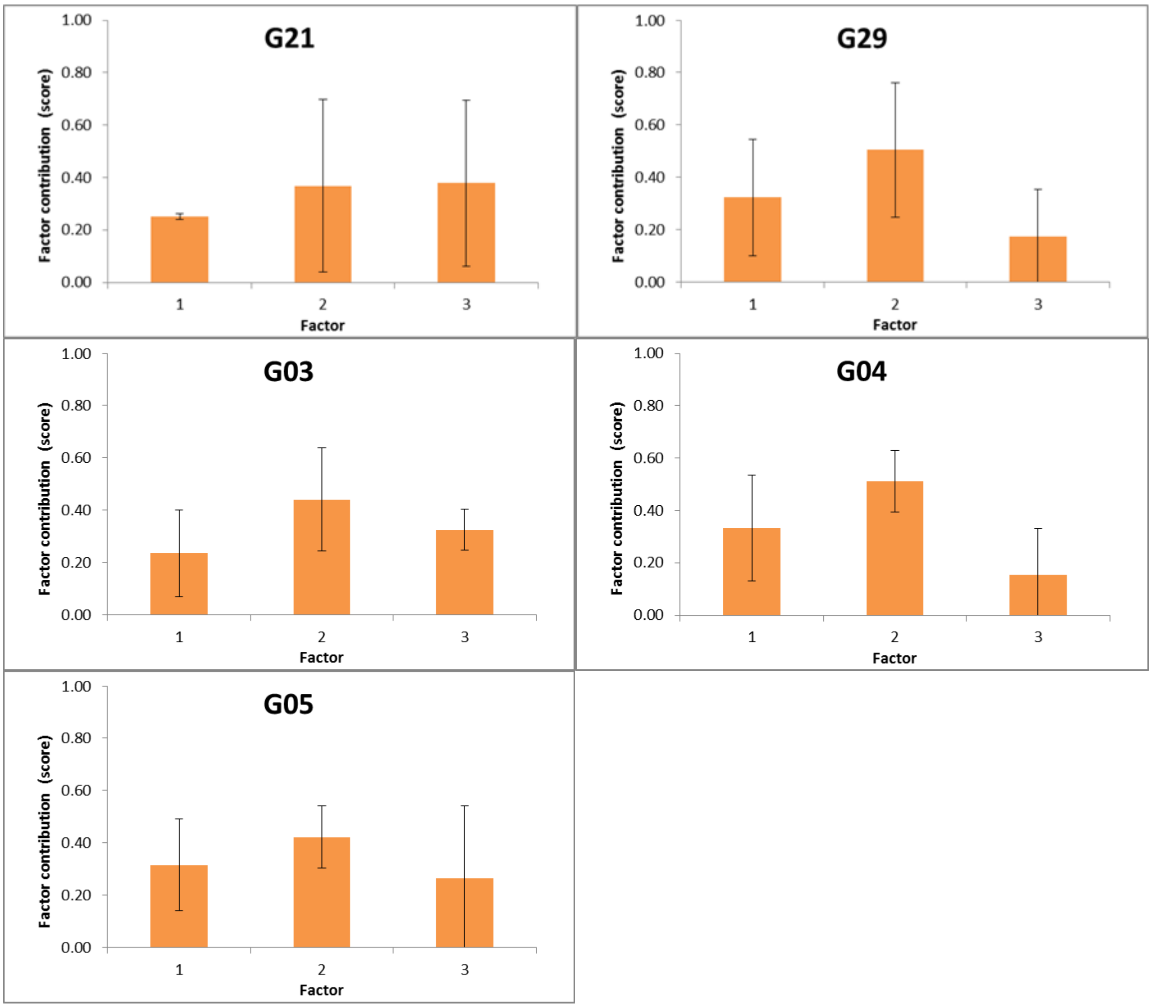


*Figure 14 Average percentage apportionments of PAHs measured in winter for each of the measuring units of Genk. The relative factor scores indicate which proportion of each factor of the factor analysis is contributing to the PAH mixture measured on that location. Factor 1 = Wood burning&traffic, Factor 2 = Traffic or?, Factor 3 = Natural gas combustion?*

### Menen

Menen showed rather similar factors in summer-fall and winter. In summer-fall the **first two factors** (together=70% of total variance) pointed at vehicle emissions. The **third factor** (14% of total variance) had relatively high factor loadings for BaA, BbF, BeP, BaP, and also CHR to a lower extend. Ravindra et al. (2008) and Callen et al. (2014) assigned high factor loadings of BaA, BbF and BaP to gasoline, vehicle emissions, which could however also originate from wood, coal and light oil combustion (Callen et al., 2014; Teixeira et al., 2013).

In winter LEV was significantly ($P<0.05$) correlated with most lower molecular PAHs. Part of **factor 1** (factor 1= 52% of total variance) could be explained by wood combustion. As for Genk also in Menen another part of factor 1 might be explained by vehicle emissions. The **second factor** (19% of total variance) was loaded with the 5- and 6-ring PAHs was also similar to the summer period and the one in Genk with the main source identified as vehicle emissions, but might also have other origines. The **third factor** with relatively high factor loadings for BeP, BaP, as in summer-fall, might be assigned to combustion or industry?

*Table 25: Factor loadings for Menen in both sampling seasons, obtained via PCA analysis (factor loadings > 0.7 are indicated in red, those above 0.43 indicated in blue)*

| **Menen** | **Summer-fall (Sept'15)** | | | **Winter (Jan-Feb'16)** | | |
|---|---|---|---|---|---|---|
| | **Factor1 Traffic (LMW PAH)** | **Factor2 Traffic and/or?** | **Factor3 Industry & combustion products** | **Factor1 Wood burning &traffic(LMW PAH)** | **Factor2 Traffic and/or?** | **Factor3 Industry & combustion products** |
| **NAP** | 0.86 | -0.30 | 0.22 | 0.88 | 0.13 | 0.36 |
| **2MeNAP** | 0.96 | 0.06 | 0.06 | 0.87 | 0.18 | 0.36 |
| **1MeNAP** | 0.94 | -0.01 | 0.08 | 0.86 | 0.21 | 0.37 |
| **ACY** | 0.71 | 0.00 | 0.08 | 0.08 | 0.18 | 0.41 |
| **ACE** | 0.91 | 0.01 | 0.10 | 0.91 | 0.12 | 0.25 |
| **FLU** | 0.97 | 0.09 | 0.01 | 0.88 | 0.17 | 0.39 |
| **PHE** | 0.97 | 0.05 | 0.07 | 0.87 | 0.12 | 0.43 |
| **9MePHE** | 0.43 | -0.64 | 0.44 | 0.72 | -0.12 | 0.40 |
| **ANT** | 0.89 | -0.31 | 0.27 | 0.75 | 0.20 | 0.55 |
| **2MeANT** | 0.62 | -0.32 | 0.23 | 0.82 | 0.21 | 0.47 |
| **FLA** | 0.92 | 0.12 | 0.28 | 0.94 | 0.11 | 0.24 |
| **PYR** | 0.78 | -0.02 | 0.30 | 0.93 | 0.13 | 0.30 |
| **BaA** | 0.22 | -0.28 | 0.81 | 0.93 | 0.05 | -0.11 |
| **CHR** | 0.46 | 0.55 | 0.57 | 0.93 | 0.15 | 0.03 |
| **BbF** | 0.55 | 0.04 | 0.79 | 0.77 | 0.29 | 0.51 |
| **BkF** | -0.28 | 0.84 | 0.40 | 0.64 | 0.18 | 0.50 |
| **BeP** | 0.10 | 0.22 | 0.85 | 0.39 | 0.22 | 0.82 |
| **BaP** | 0.10 | 0.15 | 0.94 | 0.54 | 0.11 | 0.76 |
| **IcdP** | -0.27 | 0.77 | 0.40 | -0.12 | -0.99 | -0.09 |
| **DahA** | -0.12 | 0.74 | 0.27 | -0.12 | -0.99 | -0.09 |
| **PIC** | 0.28 | 0.92 | -0.06 | -0.12 | -0.99 | -0.08 |
| **BghiP** | 0.17 | 0.93 | -0.18 | -0.12 | -0.99 | -0.09 |
| **LEV** | - | - | - | 0.81 | -0.08 | -0.03 |
| **Eigenvalue** | **0.43** | **0.21** | **0.19** | **12.02** | **4.41** | **3.62** |
| **% Variance** | **0.47** | **0.23** | **0.14** | **0.52** | **0.19** | **0.16** |

*LMW: low molecular weight*

The relative factor scores of each of the measuring units in Menen indicate which proportion each factor of the factor analysis is contributing to the PAH mixture measured on that location. At the industry locations M01 and M02 there was an even contribution of the traffic and industry factors. The background stations M03 and M04 showed somewhat similar patterns, with M04 being influence mostly by factor 3 ('industry & combustion products'). The traffic stations M05 and M06 clearly were influenced by the low molecular weight traffic related PAHs of factor 1 (Figure 15). In winter the industry location M11 showed high molecular weight PAHs (IcdP, DahA, PIC and BghiP) included in factor 2 ('traffic and/of?'). The industry location M02 and the traffic location M06 had mainly PAH immissions related to the 'industry and combustion products' factor 3. Whereas in the background stations M03 and M05 traffic and wood burning PAHs were dominating (factor 1) (Figure 16).

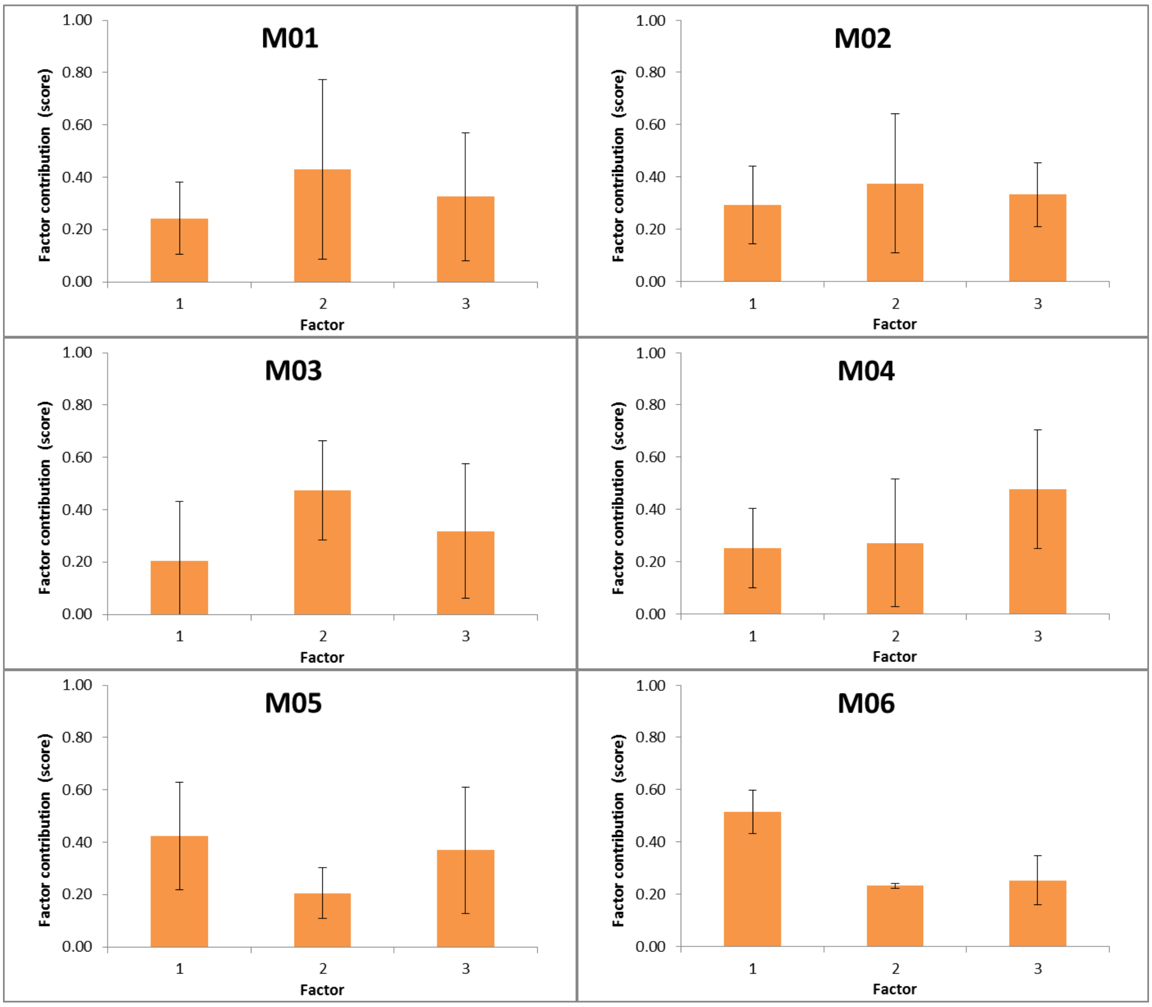


*Figure 15 Average percentage apportionments of PAHs measured in summer-fall for each of the measuring units of Menen. The relative factor scores indicate which proportion of each factor of the factor analysis is contributing to the PAH mixture measured on that location. Factor 1 = Traffic (LMW PAH), Factor 2 = Traffic and/or?, Factor 3 = Industry & combustion products.*

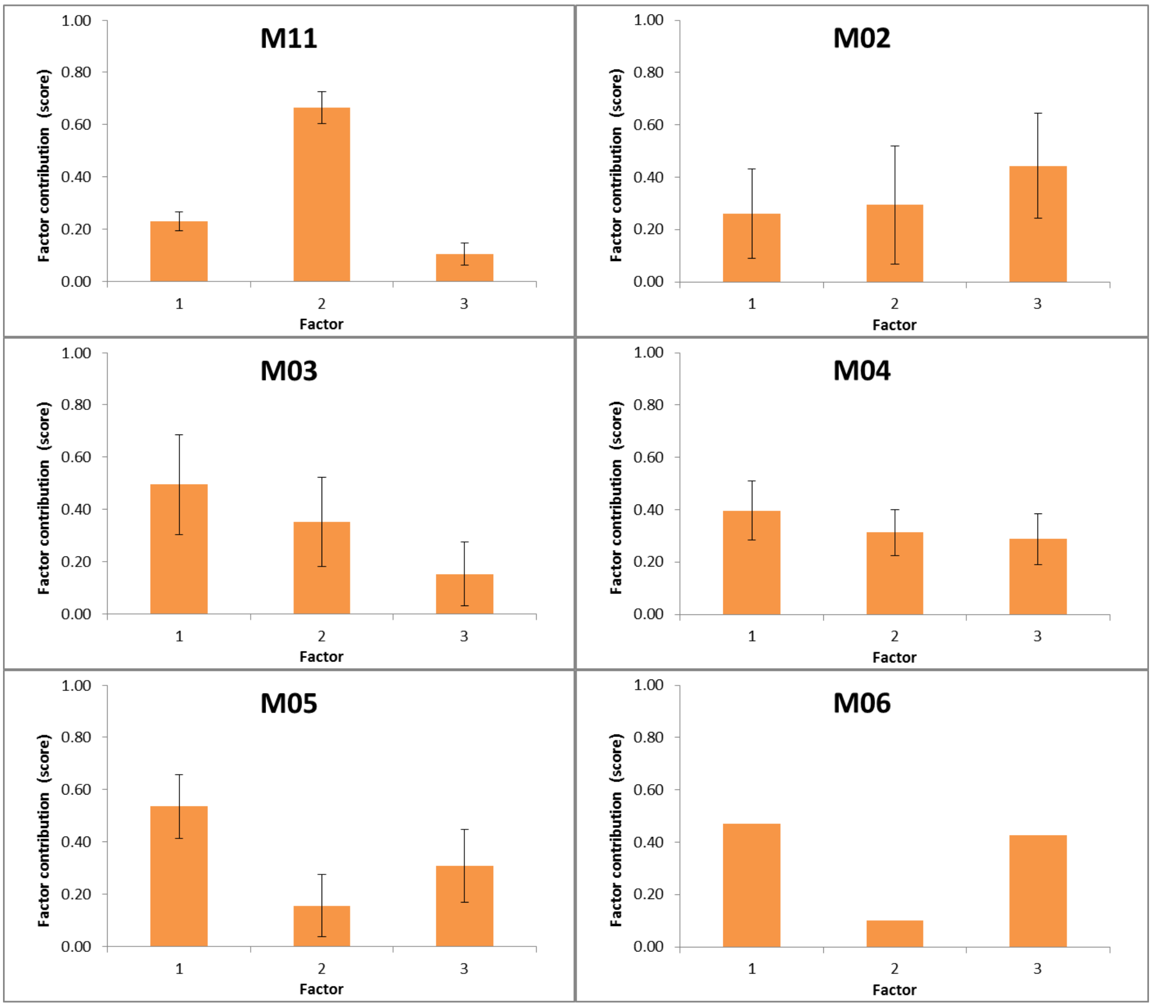


*Figure 16: Average percentage apportionments of PAHs measured in winter for each of the measuring units of Menen. The relative factor scores indicate which proportion of each factor of the factor analysis is contributing to the PAH mixture measured on that location. Factor 1 = Wood burning &traffic(LMW PAH), Factor 2 = Traffic and/or?, Factor 3 = Industry & combustion products.*

The principal component source apportionment was also checked by correlating the average of each PAH component (for each of the measuring units), with geographical information that was available from these locations. It was clearly seen, that in Genk/Diepenbeek the levels of NAP, ANT, 2MeANT, CHR, FLU, PHE, FLA, and PYR were significantly correlated with the traffic intensity on the nearest road. 9MePHE was correlated with heavy traffic intensities on the nearest road. The compound ACY was associated with the surface of industry or commercial units within a 3000m perimeter (Table 26, Figure 17). In Menen, 1MeNAP and 9MePHE were correlated with the above mentioned industry parameter. These compounds were also correlated with nearby heavy traffic intensities (Figure 17). BeP and BaP of factor 3 in the factor analysis (Table 25), appeared to be non-significantly correlated with nearby industry (Table 26)

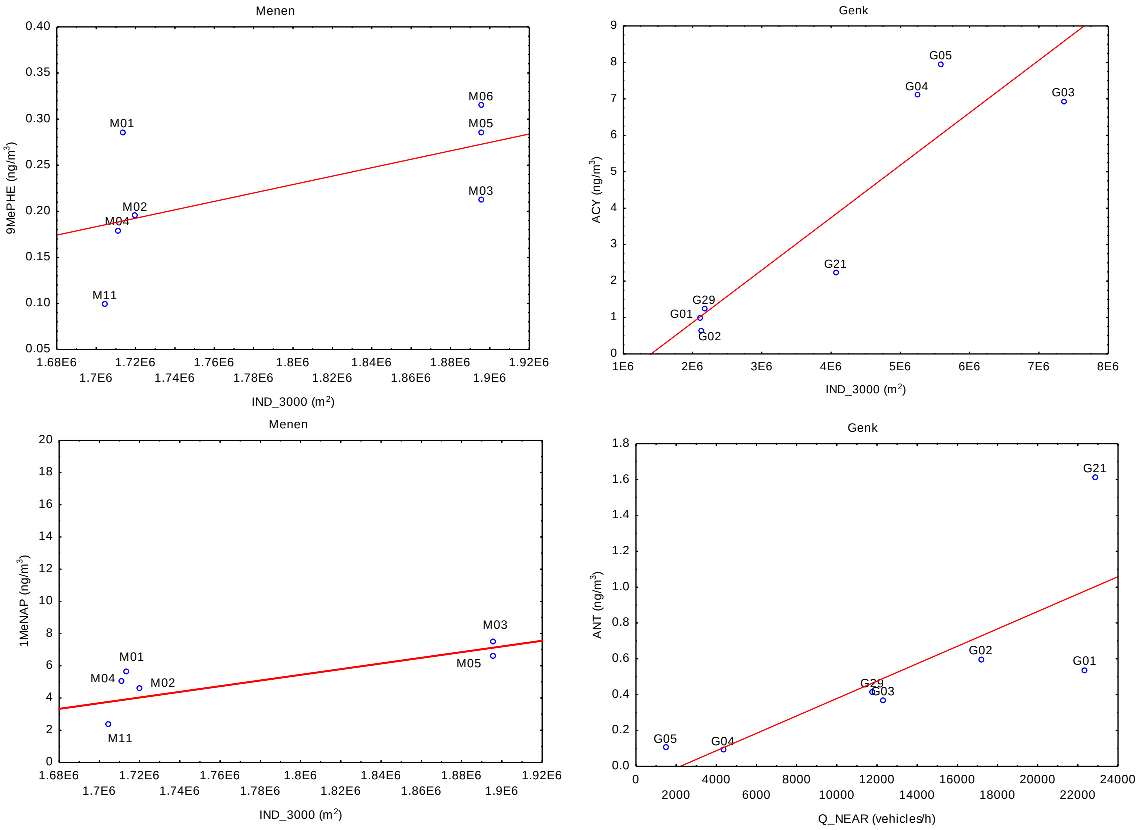


Figure 17: Scatter plot of geographical parameters versus average PAH levels observed (over both seasons) at the measuring stations in Menen or Genk (IND_3000: Industrial or commercial units in a buffer with size 3000m (CORINE Class 121) [m²]; Q_NEAR: traffic intensity on the nearest road [veh/day])

*Table 26: Spearman rank correlation coefficients between PAH or LEV levels (averaged for both season over each of the measuring units) and geographical information on habitation, industry and traffic intensities in the near of the measuring units. Significant correlations are indicated in red ($p<0.05$).*

| | **Genk/Diepenbeek** | | | | | | | **Menen** | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| PAH | **ADDRESS_1000** | **IND_3000** | **NATURE_5000** | **Q_NEAR** | **D_NEAR_MAJOR1** | **Q_NEAR_HEAVY** | **busstops_500** | **ADDRESS_1000** | **IND_3000** | **NATURE_5000** | **Q_NEAR** | **D_NEAR_MAJOR1** | **Q_NEAR_HEAVY** | **busstops_500** |
| **NAP** | -0.14 | -0.25 | 0.14 | 0.86 | 0.61 | 0.70 | 0.45 | -0.04 | 0.22 | -0.43 | 0.61 | 0.36 | -0.14 | 0.15 |
| **2MeNAP** | -0.04 | -0.57 | 0.43 | 0.75 | 0.61 | 0.67 | 0.38 | 0.14 | 0.33 | -0.54 | 0.67 | 0.54 | -0.02 | 0.35 |
| **1MeNAP** | -0.79 | 0.46 | 0.50 | 0.32 | 0.00 | 0.44 | -0.18 | 0.64 | 0.85 | -0.29 | 0.41 | 0.54 | 0.74 | 0.47 |
| **ACY** | -0.57 | 0.86 | 0.11 | -0.68 | -0.96 | -0.48 | -0.88 | 0.04 | 0.70 | -0.54 | -0.05 | 0.14 | -0.02 | 0.42 |
| **ACE** | -0.07 | 0.14 | 0.07 | -0.36 | -0.46 | -0.37 | -0.59 | 0.04 | 0.30 | -0.25 | 0.50 | 0.29 | -0.14 | 0.22 |
| **9MePHE** | -0.29 | -0.39 | 0.68 | 0.64 | 0.57 | 0.82 | 0.16 | 0.54 | 0.82 | 0.00 | 0.25 | 0.29 | 0.61 | 0.35 |
| **ANT** | 0.18 | -0.61 | 0.07 | 0.89 | 0.79 | 0.67 | 0.67 | -0.11 | 0.15 | -0.29 | 0.07 | 0.07 | -0.43 | 0.18 |
| **2MeANT** | 0.07 | -0.54 | 0.18 | 0.86 | 0.82 | 0.70 | 0.59 | -0.07 | 0.37 | -0.21 | 0.40 | 0.14 | -0.11 | 0.13 |
| **FLU** | 0.18 | -0.61 | 0.07 | 0.89 | 0.79 | 0.67 | 0.67 | 0.11 | 0.22 | -0.36 | 0.54 | 0.43 | 0.00 | 0.33 |
| **PHE** | 0.11 | -0.64 | 0.21 | 0.93 | 0.75 | 0.78 | 0.63 | -0.07 | 0.11 | -0.25 | 0.49 | 0.25 | -0.13 | 0.13 |
| **FLA** | 0.18 | -0.61 | 0.07 | 0.89 | 0.79 | 0.67 | 0.67 | -0.14 | 0.19 | -0.14 | 0.45 | 0.11 | -0.27 | 0.02 |
| **PYR** | 0.18 | -0.61 | 0.07 | 0.89 | 0.79 | 0.67 | 0.67 | -0.11 | 0.30 | -0.32 | 0.58 | 0.21 | -0.29 | 0.04 |
| **BaA** | -0.18 | -0.36 | 0.39 | 0.75 | 0.57 | 0.70 | 0.20 | -0.14 | 0.44 | 0.25 | -0.14 | -0.36 | 0.04 | -0.16 |
| **CHR** | -0.29 | -0.29 | 0.43 | 0.82 | 0.46 | 0.78 | 0.13 | -0.43 | 0.22 | -0.11 | 0.00 | -0.25 | -0.40 | -0.07 |
| **BbF** | -0.18 | -0.36 | 0.39 | 0.75 | 0.57 | 0.70 | 0.20 | -0.50 | 0.22 | -0.14 | 0.04 | -0.29 | -0.47 | -0.16 |
| **BkF** | -0.11 | -0.39 | 0.39 | 0.64 | 0.57 | 0.70 | 0.20 | -0.21 | 0.44 | 0.21 | -0.11 | -0.39 | -0.04 | -0.25 |
| **BeP** | -0.04 | -0.36 | 0.32 | 0.46 | 0.46 | 0.56 | 0.11 | -0.14 | 0.44 | 0.25 | -0.14 | -0.36 | 0.04 | -0.16 |

| PAH | Genk/Diepenbeek | | | | | | | Menen | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | **ADDRESS_1000** | **IND_3000** | **NATURE_5000** | **Q_NEAR** | **D_NEAR_MAJOR1** | **Q_NEAR_HEAVY** | **busstops_500** | **ADDRESS_1000** | **IND_3000** | **NATURE_5000** | **Q_NEAR** | **D_NEAR_MAJOR1** | **Q_NEAR_HEAVY** | **busstops_500** |
| **BaP** | -0.04 | -0.36 | 0.32 | 0.46 | 0.46 | 0.56 | 0.11 | -0.14 | 0.44 | 0.25 | -0.14 | -0.36 | 0.04 | -0.16 |
| **IcdP** | 0.25 | -0.43 | 0.21 | -0.07 | 0.32 | 0.04 | 0.02 | -0.64 | -0.19 | 0.64 | -0.38 | -0.89 | -0.56 | -0.82 |
| **DahA** | 0.11 | -0.39 | 0.32 | 0.07 | 0.29 | 0.19 | -0.09 | -0.71 | -0.33 | 0.61 | -0.56 | -0.93 | -0.52 | -0.76 |
| **BghiP** | 0.07 | -0.36 | 0.29 | 0.18 | 0.32 | 0.30 | -0.04 | -0.54 | -0.37 | 0.25 | 0.32 | -0.36 | -0.72 | -0.67 |
| **PIC** | 0.11 | -0.39 | 0.32 | 0.07 | 0.29 | 0.19 | -0.09 | -0.29 | -0.41 | 0.57 | 0.31 | -0.32 | -0.59 | -0.56 |
| **LEV*** | 0.03 | -0.20 | 0.37 | 0.03 | 0.43 | 0.09 | 0.35 | -0.36 | -0.19 | -0.25 | -0.81 | -0.32 | -0.13 | 0.09 |

ADDRESS_1000: number of addresses in a buffer with size 1000; IND_3000: Industrial or commercial units in a buffer with size 3000m (CORINE Class 121) [m²]; NATURE_5000: natural land in a buffer with size 5000m (CORINE Classes 141, 142, 211, 231, 242, 243, 311, 312, 313, 322, 421, 511, 512, 522) [m²]; Q_NEAR: traffic intensity on the nearest road [veh/day]; D_NEAR_MAJOR1: 1 / distance to the nearest major road [1/m]; Q_NEAR_HEAVY: traffic intensity (heavy traffic) on the nearest road [veh/day]; busstops_500: number or busstops in a buffer with size 500m

* correlation calculated for levoglucosan levels measured in winter season

## 3.6. INFORMATION ON PAH INTAKE VIA FOOD VIA FOOD QUESTIONNAIRE

The participants of the study had to fill out a food frequency questionnaire. Consumption of possible PAH containing food items were asked for. The median consumption of cereals (including bread, sandwiches, muesli) was 112 g/day (0-350 g/day, min-max), and 10.0 g/day for mayonnaise and oils (0-120 g/day, min-max). Participants reported drinking 275 mL coffee per day (median, with min-max: 0-887 mL).

Li et al. (2010) advised for studies on non-dietary sources, to avoid consumption of smoked, grilled, and barbecued food or even use it as an exclusion criterion to prevent data biasing. Recently, Rose et al. (2015) reported little evidence of PAH formation during grilling, frying and toasting experiments, whereas barbecuing over charcoals gave increased PAHs in food. In the current study, the participants were asked to avoid grilled, roasted, and barbecued food. From the 383 urine samples collected, respectively three cases were reported of barbecue or burnt/grilled food consumption 24h before the urine sampling. These urine samples contained: 238.8, 38.4 and 28.2 ng/g CRT 1-OHPyr, and respectively 1979.2, 637.6 and 541.6 ng/g CRT OH-phenanthrenes. Meaning that the first person had levels well beyond the average, which was not observed in the other two individuals.

The participants were allowed to eat smoked food, and reported their 24h consumption of smoked cheese, fish and meat products. We did not observe a clear (significant) trend in impact of this consumption on the 1-OH pyrene levels in urine (
Figure 18).

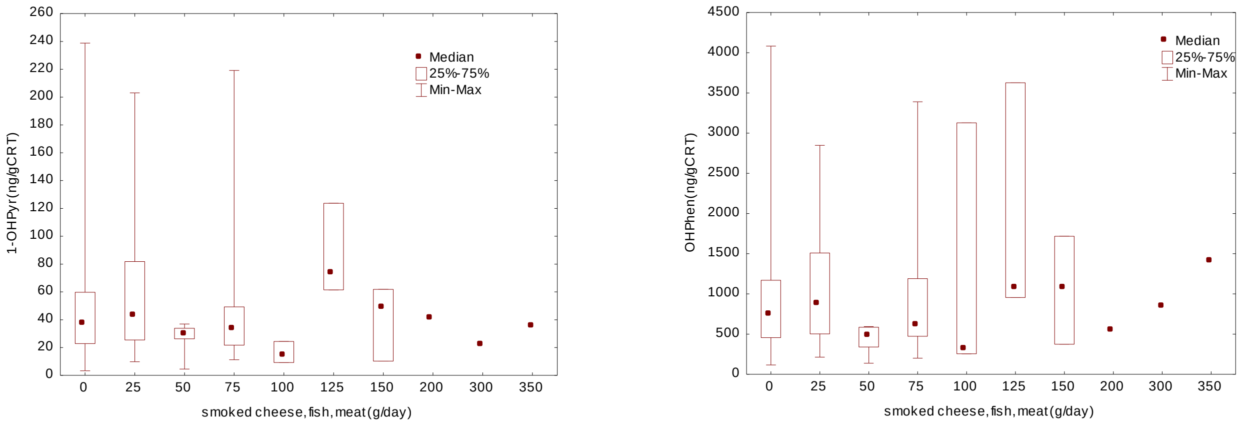


*Figure 18: 1-OH pyrene and OH-phenanthrene urine levels (ng/g CRT) observed in each of the 363 urine samples collected over the current campaign, presented for the different amounts of smoked food consumed 24h before urine collection (classes: no consumption, between 0 and 25, between 25 and 50,…between 300 and 350, and >350 g/day)*

## 3.7. URINARY LEVELS OF HYDROXY-PAH METABOLITES

The median concentration of OH metabolites was respectively 35.5 ng/L (Genk) vs. 37.7 ng/L (Menen) for 1-OH pyrene and 615.9 ng/L (Genk/Diepenbeek) vs. 717.8 ng/L (Menen) for the sum of OH-phenanthrenes (Table 27).

It was earlier observed that at age above 60y, PAH metabolite levels were lower compared to the concentrations in younger adults (Thai et al., 2016). Therefore we need to compare the urinary levels with a similar age group of 50-65 y old adults monitored during the FLEHS-1 human biomonitoring study performed all over Flanders (De Coster et al., 2008). In the current study population, respectively 14, 44 and 42% of the individuals were of the age group between 33-49y, 50-60y, or >60y. We did not see a trend in the PAH urinary metabolite levels in this age range (Figure 19), therefore we compared the across-ages median levels with the Flemish (FLEHS-1) adult population. The overall median 1-OH pyrene urinary level in the FLEHS-1 group was 79 ng/L (interquartile range = 30-195 ng/L) (N=1575), which was considerably higher than the levels observed in the current study. However, the current study population was asked to avoid certain food items 24h before urine collection, were non-smokers, were not allowed to have wood or coal stoves in their residences, and were asked not to use candles, or burn any essences indoor, during 24h before urine collection. When excluding from the FLEHS-1 population, individuals which either smoked, recently ate barbecued/grilled food, or had indoor heating with a stove, the median(IQR) 1-OH pyrene level of the Flemish population was 30 (30-140) ng/L, which was in the range of the concentrations measured in the current campaign. Similar 1-OH pyrene levels were reported in the same age classes (smokers+non-smokers) in: USA 32.5 ng/L (N=507) (Best et al. 2016), and Australia where urine pools were analysed with levels between 50 and 96 ng/L (Thai et al., 2016). In the latter study, the urine pools of participants between 15 and 29 years old had 1-OH pyrene levels of 190 and 270 ng/L, which was comparable with the adolescents' urinary concentrations earlier measured in Menen and Genk/Diepenbeek.

*NB: In the FLEHS-1 adult population the median 1-OH pyrene urine concentration in non-smokers 63(30-150)ng/L (N=1261) was considerably lower that the levels observed in smokers: 276(114-591)ng/L (N=276). Furthermore, non-smoking individuals having consumed barbecued food within three days before urine collection had median levels of 81(30-167)ng/L (N=40), whereas the non-smokers not having consumed barbecued food within that time showed levels of 60(30-147)ng/L (N=1116). Also among the non-smokers,* *individuals having a stove for heating, or having reported indoor burning of wood, paper, or household rests, had higher levels of 1-OH pyrene: 68(30-144) (N=305 with stove) vs. 62(30-155) (N=877 without stove), and 69(30-155)ng/L (N=496 burning indoors) vs. 30 (30-147)ng/L (no burning indoors).*

*Table 27: Urinary levels of 1-OH pyrene and sum of OH-phenanthrenes in all samples collected*

| | N | Mean | Geometric mean | Med | Min | Max | $P_{25}$ | $P_{75}$ | St.Dev. |
|---|---|---|---|---|---|---|---|---|---|
| **Genk/Diepenbeek** | | | | | | | | | |
| **CRT** (mg/dL) | 162 | 114.6 | 103.9 | 115.0 | 17.0 | 258.0 | 77.0 | 150.0 | 46.4 |
| **1-OHPyr**(ng/L) | 160 | 39.2 | 31.9 | 35.5 | 3.3 | 143.0 | 22.5 | 48.1 | 25.1 |
| **OHPhen**(ng/L) | 160 | 730.1 | 635.3 | 615.9 | 155.1 | 2852.8 | 449.9 | 872.5 | 428.1 |
| **1-OHPyr**(ng/gCRT) | 160 | 41.2 | 30.8 | 33.4 | 3.3 | 238.8 | 21.2 | 49.0 | 35.8 |
| **OHPhen**(ng/gCRT) | 160 | 796.6 | 613.3 | 560.8 | 116.1 | 3846.6 | 401.9 | 925.1 | 690.6 |
| **Menen** | | | | | | | | | |
| **CRT** (mg/dL) | 165 | 97.7 | 86.4 | 91.0 | 19.0 | 296.0 | 66.0 | 128.0 | 46.6 |
| **1-OHPyr**(ng/L) | 164 | 43.3 | 35.4 | 37.7 | 2.7 | 252.8 | 25.6 | 53.0 | 30.9 |
| **OHPhen**(ng/L) | 164 | 918.7 | 770.2 | 717.8 | 250.3 | 4580.2 | 523.8 | 1106.4 | 651.6 |
| **1-OHPyr**(ng/gCRT) | 164 | 52.0 | 41.1 | 45.3 | 6.0 | 225.3 | 28.4 | 67.0 | 37.1 |
| **OHPhen**(ng/gCRT) | 164 | 1106.8 | 893.9 | 889.8 | 205.4 | 4082.1 | 550.5 | 1438.8 | 768.1 |

The levels of 1-OH pyrene and sum of the OH-phenanthrenes were correlated with a Spearman rank correlation coefficient of r=0.57 (Figure 19). This correlation was also observed in other studies, such as performed in the USA (CDC, 2015), United Kingdom (Aquilina et al., 2010), and Canada (Health Canada, 2013). It supports the general use of 1-OH pyrene as a representative biomarker for overall PAH exposure (Thai et al., 2016).

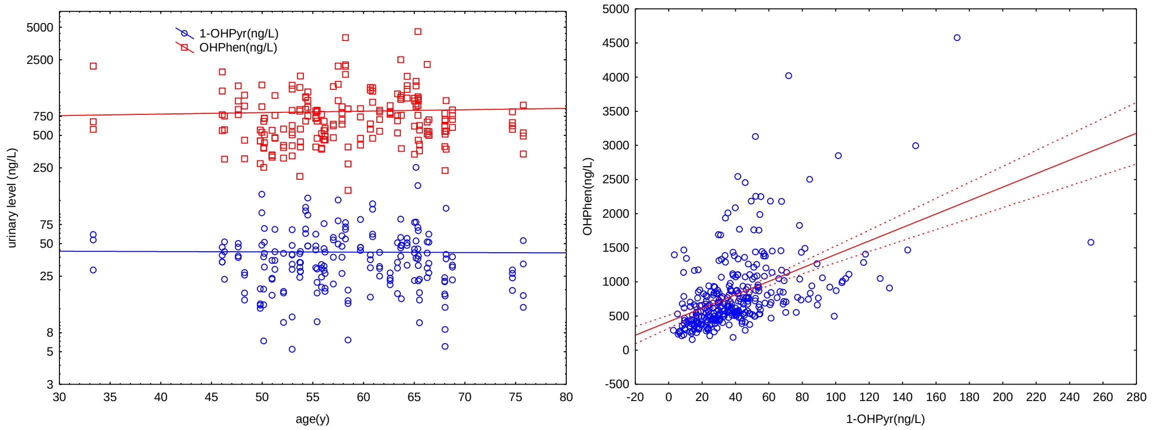


*Figure 19: 1-OH pyrene and OH-phenanthrene levels in relation to age of the participants (left). Correlation of 1-OH pyrene with OH-phenanthrene urinary levels (right).*

## 3.8. RELATIONSHIP OF AIR PAH AND LEVOGLUCOSAN LEVELS AND URINARY CONCENTRATIONS

The variables that were tested as determinants of the urinary PAH metabolites 1-OH pyrene and sum of OH-pyrenes were: passive smoking, sex, age, body mass index (BMI), highest education within family, food intake of possible PAH containing products, number of years living in area, region of living, relevant geographical information on the living environment in the context of PAH exposure, type of indoor heating, type of cooking system, working outside community, being underway in busy traffic, garage connected to residence.

Based on mixed effects single linear regression models, only sex showed a potential influence on creatinine adjusted 1OH-pyrene levels. Average 1OH-pyrene levels were 1.46 times higher in women as compared to men (p-value 0.02). For sum of urinary OH-phenanthrenes, the variables being determinants were region, parents living in Belgium, and coffee consumption. In Menen, the average OH-phenanthrene levels were 1.36 times higher as compared to Genk (p-value 0.01) (Figure 20). Individuals from which the parents were born in Belgium, had 1.84 times higher levels than those who were not (p=0.001). There was also a minor, but significant increase of OH phenanthrenes in urine, of 0.06% per doubling of intake of coffee intake (p=0.009) (Table 28).

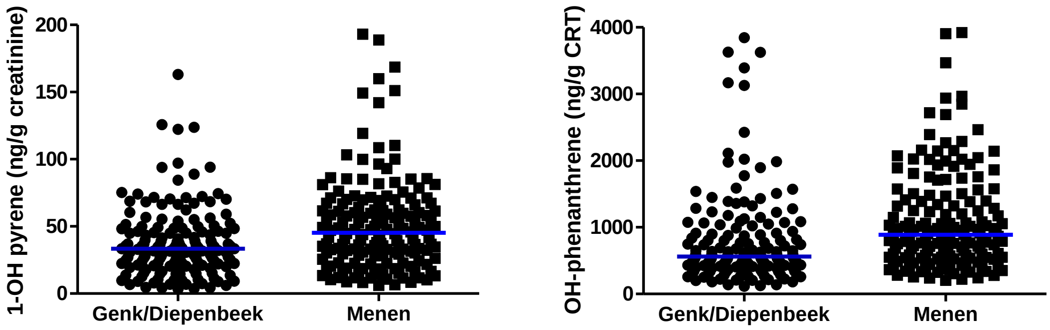


*Figure 20: Urinary 1-OH pyrene and OH phenanthrene concentrations of all collected urine samples in Genk/Diepenbeek and Menen. The blue line represents the median levels in both regions.*

After adjustment for the above mentioned covariates there was no significant association observed between ambient PYR or levoglucosan concentrations and 1-OH pyrene in urine. In the same way, also ambient PHE+ANT or levoglucosan concentrations were not correlated with OH-phenanthrenes in urine. Aquilina et al. (2010) reported no correlation between FLU, PHE or PYR measured via personal samplers and 1-OH pyrene analysed in urine of 100 non-smoking subjects living in the UK. In a study on non-smoking pregnant women, high density housing, passive smoking exposure, and residential heating with coal showed up as significant predictors of PAH exposure (Polanska et al., 2014). However the influence was not seen on all OH-metabolites in urine, which made them concluding that assessing four OH-phenanthrene metabolites (2-OH, 3-OH and 9-OH-PHE, as well as PHE-9,10 diol) allows characterization of environmental PAH exposure.

*Table 28: Result of the mixed effects single linear regression models in which the factors of influence on the urinary levels of 1-OH pyrene and OH phenanthrenes were examined*

| Covariates | Estimate | SE | CI $_{under}$ | CI $_{upper}$ | p |
|---|---|---|---|---|---|
| **1-OH pyrene** | | | | | |
| Sex (women compared to men) | 1.44 | 1.17 | 1.06 | 1.95 | 0.02 |
| **OH phenanthrenes** | | | | | |
| Living in Menen (compared to Genk/Diepenbeek) | 1.37 | 1.13 | 1.07 | 1.75 | 0.01 |
| Parents born in Belgium (yes) | 1.06 | 1.02 | 1.03 | 1.10 | 0.001 |
| coffee consumption (mL/day) | 1.0006 | 1.0002 | 1.0002 | 1.0011 | 0.008 |

CI$_{under}$ and CI$_{upper}$: under and upper limit of the 95% confidence interval of the estimate

# CHAPTER 4 COMMUNICATION WITHIN THE STUDY

At the start of the project local ambassadors and local committee members were active in recruiting people interested in joining the study. The major of each of the communities was informed about the start and the content of the study via a letter. The first contact with possible participants was made by the local actors. The study team indicated the streets, which the participants needed to be recruited in. The addresses of the people willing to participate were gathered by the local actors and were communicated to VITO and PIH. The latter took up further contact to arrange the sampling dates, and all possible practical arrangements. Right after both of the seasonal campaigns, the participants got an e-mail informing the number of participants, the number of samples collected, and the analyses that would be performed on the samples.

The current report is part of the public communication. Furthermore, the participants obtain a letter with all their personal results, and a short communication on the major results of the study. Both in Menen and Genk/Diepenbeek the local actors - including representatives of the community, the local environment and health workers, representatives of industry – are informed via an oral presentation and discussion on the results. Aim is to get some understanding of the results, and possible local actions that could be formulated.
On the Flemish level, a discussion among the different administrations involved in environment was started to formulate advices to the Flemish Minister of Environment considering traffic and wood burning policy.

# CHAPTER 5 CONCLUSION AND POLICY RECOMMENDATIONS

Based on increased 1-OH pyrene levels in Menen and Genk/Diepenbeek, observed during the FLEHS-3 biomonitoring, the current study was started to assess possible local sources of outdoor PAHs in those regions. To assess PAH and levoglucosan exposure, 13 measuring units were set up near to habitation in the backyard or on balcony of different residences in those regions.

The PAH concentration measured in Menen and Genk/Diepenbeek were in the range of what was earlier measured in other European regions. Clearly, Menen and Genk/Diepenbeek had higher concentrations for most of the PAHs compared to the background location Houtem, although this difference was much less apparent in the winter period. Lower molecular weight PAHs were correlated with levoglucosan levels measured at the same locations, indicating that biomass burning was an important source of that type of PAHs in all regions. Indeed in colder periods, levoglucosan increased consistently on all measuring locations. Overall, the levels of levoglucosan observed were comparable to what was earlier measured in larger campaign done in Flanders (Maenhaut et al. ,2012). The PAH profiles on most of the measuring locations indicated that traffic was one of the important sources generating PAHs in the residences ambient air environment. Traffic counts at the measurement location would have been very useful to assess the real-time traffic pressure at the home locations.

We could not observe a link between ambient PAH concentrations and urinary OH-metabolites repeatedly measured in the participants, in each of the air sampling weeks. Since intake of PAHs via smoking and food consumption were excluded or reduced to a minimum via the study design, PAH levels in ambient air were beforehand speculated to be the main cause for concentration variations seen within the individuals. Why, then there was no observed link between air PAHs and 1-OH pyrene or OH-phenanthrenes? Aside from exposure on the home address, individuals also accumulated PAHs indoors and on other than home locations. The time frame of internal metabolisation did not completely overlap with the time frame of the 48-hours air sampling. Indeed, the half-life of PAH metabolites depends on the compound and certainly also on the exposure route, being: 6.0-29.0 h after inhalation exposure; 4.4-12h or 3.1-5.5h after ingestion exposure, and 11.5-15h for dermal absorption (numbers cited in and reported by Li et al., 2012). It can be noted, that ideally PAH OH-metabolite levels of different age groups are measured, as metabolisation and exposure patterns (food, ambient/occupational/hobby) differ among different ages (Thai et al., 2016). As lower molecular weight PAHs are the main PAHs present in the air of people’s life environment, it may be interesting to assess mainly these compounds as proxy for PAH exposure. Certainly 1-OH pyrene may lack some precision to assess the PAH exposure pattern observed in real life.

PAHs were ubiquitously present on all locations monitored in the current study. It shows that analysis of environmental concentrations is useful, as a whole profile of compounds can be assessed and some indication can be obtained on possible sources. To identify PAH exposure of a population or community, several measuring locations should be set up in order to monitor different local sources (industrial sources, differences in traffic intensity, differences in traffic composition, presence other combustion sources). Indeed, the stations in a same community in the current study varied considerably in profiles even if they were located within perimeter of max 2-3 km. They could not be used to assess the diversity of PAH exposure in a wider area.

Measurements of levoglucosan showed to be less subject to local variation, but were mainly influenced by temporal (temperature) variations. The winter levels often showed to be representative of a wider region; i.e. being similar on other measuring locations.

**Measures for follow-up and reduction of PAH exposure**

Air contamination with PAHs remains a matter of concern in whole over Europe and beyond. PAHs are emitted via residential heating, traffic, an industry. Initiatives on personal exposure prevention, and local or regional measures should be stimulated.

*Minimizing personal exposure to PAHs*

We assessed personal exposure, measuring the urinary markers 1-OH pyrene and OH-phenanthrenes. As mentioned above, the population of this study was clearly selected, avoiding personal exposure to PAHs other than via ambient air: older population not occupationally exposed to PAHs, non-smokers, no indoor stoves, open fires or candle burning, no grilled/barbecued food consumption in the last 24h before urine collection. These are the main life-style factors that determine PAH exposure. These are at the same time the best measures that can be taken to avoid personal exposure. Indeed, in the current study the levels of 1-OH pyrene were low: 35.5 (Genk) and 37.7 (Menen). However, some of the factors, such as the use of stoves does not necessarily imply increased PAH levels indoors and outdoors in the neighborhood, when this is done in a good way. Advices to citizens are important in this context. Citizens can change their behavior to obtain lower exposure to PAHs by avoiding sources of PAHs, such as: no smoking, avoiding consumption of burned food, maintenance and use of good ventilation systems, smart ventilation and airing (orientation of ventilation systems and open windows away from local sources, opening windows during periods with low traffic activity), application of best practice for barbecuing of food, apply best practice for the use of woodstoves.

The Flemish authorities can offer support by investing in informing citizens and raising awareness about best practices. Several efforts on these topics were already initiated and this study emphases the importance of the continuation and possible elaboration of these campaigns. Some examples:

- Ban on open fires and waste burning
- Best practice for use of (wood)stoves: stook slim (https://www.lne.be/campagnes/stook-slim)
- Best practice for ventilation and airing (http://www.vlaanderen.be/nl/publicaties/detail/ventileren-en-verluchten)
- Awareness raising on smoking (http://www.gezondheidenmilieu.be/nl/projecten/actieweek_gezond_binnen_2016/campagne_binnen_roken_is_nooit_oke-2045.html)
- Awareness raising healthy indoor environment (http://www.gezondheidenmilieu.be/nl/projecten/actieweek_gezond_binnen_2016-1673.html)Best practice for barbecue (http://www.kanker.be/alles-over-kanker/aantoonbaar-risico/tips-om-gezonder-te-barbecue-n, http://www.gezondheid.be/index.cfm?fuseaction=art&art_id=573)

*Minimizing PAH exposure via local measures*

Measures that can be taken in specific areas to reduce PAH exposure, can be focused on traffic, as indeed traffic compounds were clearly visible in the PAH profiles measured in the current study. There is a wide variety on possible actions to reduce traffic exposure of citizens, such as measures to improve/stimulate public traffic, traffic flows, biking. To and from the center of cities and communities biking can be stimulated by good urban planning, smart city planning and by reducing traffic. Heavy truck traffic in the neighborhood of residential areas needs to be avoided. Furthermore, communication towards citizens about actual ambient air quality and effectiveness of air quality measures, can stimulate people. Industrial sites and traffic streams should be well followed-up, and direct influences of possibly elevated emissions should be openly discussed to enable reduction measures In Menen and Genk/Diepenbeek, one could stimulate a more efficient use of water ways for industrial traffic.

In several cities similar action plans were developed, which ca be used as a guidance or source of inspiration to adopt measures that are suitable for the local situation. As an example, hereby a summing-up of action plans running in some cities:

- Air quality action plan in Ghent and Ghent harbor: https://www.lne.be/themas/luchtverontreiniging/beleid/luchtkwaliteitsplan-gent
- Antwerp Masterplan 2020: http://www.vlaanderen.be/nl/mobiliteit-en-openbare-werken/wegen/masterplan-2020-voor-de-mobiliteit-en-rond-antwerpen
- Antwerp Action plan particulate matter and $NO_2$: https://www.lne.be/themas/luchtverontreiniging/nieuwactieplanantwerpen-2014-2018-goedgekeurd.pdf
- Leuven Climat Neutral plan: http://www.leuvenklimaatneutraal.be/, http://groenleuven.be/leuven_kan_zelf_luchtkwaliteit_meten,
- Utrecht air quality plan: https://www.lne.be/themas/luchtverontreiniging/presentaties/6_Utrecht_WietBaggen.pdf
- Traffic mobility plan in Genk: http://www.genk.be/Leefomgeving/Mobiliteit_en_Verkeer/Mobiliteitsplannen_Genk_Hasselt http://www.genk.be/Leefomgeving/Leefmilieu/Milieu_informatie_Genk_Zuid:31293/Het_E_missieplan

*Minimizing PAH exposure and follow-up of PAH exposure on Flemish level*

Overall, green energy generation should be stimulated to drop considerably burning processes needed for driving residential heating, engines, and industrial plants. Stimulation of electric heating using electricity generated via photovoltaic cells or wind mills, would considerably reduce PAH levels in the direct life environment of the people. In the same way, electric cars, busses, trains, and trams powered by green energy, should be stimulated.

From the current project it was clear that NAP is one of the major PAHs, that could be measured on nearly all measuring locations (from 30-40% present in total ambient PAH mixture in summer-fall, to 40-50% in winter). NAP is part of the priority PAHs in the environment and, as it is the most abundant PAH detected in ambient air, it should not be neglected in toxicological evaluation of air quality (Wauters et al., 2006). From the results, it appeared advisable to include this compound in air quality measurement campaigns.

# CHAPTER 6 FIELD WORK DOCUMENTS

The following field work documents were used in the current study:

| | |
|---|---|
| **Annex A** | Informed consent |
| **Annex B** | General questionnaire |
| **Annex C** | Instructions for collection of urine |
| **Annex D** | Questionnaire accompanied with urine collection |
| **Annex E** | Food questionnaire |
| **Annex F** | Instructions for air sampling by participants |
| **Annex G** | Scheme for sample collection by participants |

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## ANNEX A - INFORMED CONSENT

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lne.

# Geïnformeerd toestemmingsformulier: Inschatting bronnen van blootstelling aan PAK's bij inwoners van de regio's Menen en Genk-Zuid

*Studie uitgevoerd door de Vlaamse Instelling voor Technologisch Onderzoek te Mol, het PIH te Antwerpen, de universiteit van Utrecht, de universiteit Gent. De campagne wordt gefinancierd door het departement Leefmilieu, Natuur en Energie (LNE) van de Vlaamse Overheid.*

Beste,

Het Steunpunt Milieu en Gezondheid meet in opdracht van de Vlaamse overheid de aanwezigheid en de effecten van milieuvervuilende stoffen in de mens via humane biomonitoring (HBM) (http://www.milieu-en-gezondheid.be/).
In 2012 werden o.a. vervuilende stoffen in urine van ongeveer 200 jongeren (14-15 jaar) uit Genk-Zuid en de regio Menen onderzocht. In vergelijking met jongeren uit geheel Vlaanderen, werd in deze twee industriële gebieden, een hogere blootstelling aan verbrandingsproducten, of zogenoemde polycyclische aromatische koolwaterstoffen (PAK's) vastgesteld.
PAK's kunnen afkomstig zijn van zeer diverse bronnen, waaronder verbrandingsmotoren (verkeer), gebouwverwarming, afvalverbranding, maar ze kunnen ook aanwezig zijn in bv. gerookte of gegrilde voedingswaren. De huidige studie heeft als doel de oorzaak van de vastgestelde verhoging na te gaan en de voornaamste (lokale) bronnen in kaart te brengen.

**Wie kan deelnemen?**

Wij zoeken 20 personen in Menen en 20 in Genk-Zuid die bereid zijn om deel te nemen aan deze studie. De studie loopt in twee seizoenen: zomer van 2015 en winter van 2015/2016. Er zijn dus twee meetmomenten, een zomer- en een wintercampagne. De volgende selectiecriteria gelden:

- Tussen 50 en 70 jaar oud zijn (70 jaar inbegrepen)
- Niet roken en geen rokers in huis
- Geen huidige beroepsblootstelling aan PAK's (bv. werkplaatsen waar petroleum-producten worden verbrand, cokes productie, gasproductie, metaal- of staalproductie, roofing teer en asfaltverwerking of plaatsing, afvalverbrandingsinstallatie, aluminium smelters)
- Minstens 1 jaar wonen op het huidige adres
- Niet verhuizen binnen het jaar
- Bereid zijn om meetapparatuur te plaatsen aan de woning
- Niet verwarmen met (kolen, hout, gas) kachel of open haard. Partners of twee individuen die in dezelfde woning wonen, kunnen beide deelnemen aan het onderzoek als ze voldoen aan bovenstaande criteria.

## Hoe zal het onderzoek praktisch verlopen?

Bij een aantal van de deelnemers zullen luchtmetingen uitgevoerd worden aan de gevel of in de voor- of achtertuin van de woning. Daarvoor worden door een veldwerker twee meetapparaten bevestigd aan de woning. De metingen zullen over 4 weken in de periode augustus-september 2015 en 4 weken in de periode december-januari 2015/2016 lopen. We zullen vragen om de patronen met adsorbens en de filtercasette van de stof-impactor 2x per week te vervangen. We geven u daarvoor duidelijke richtlijnen.

In het begin van elke 4-weken durende campagne (zomer en winter) vult u een algemene vragenlijst in. Alle deelnemers verzamelen een staal ochtendurine in elke week van de 4-weken durende zomer- en wintercampagne. De helft van de urinestalen worden tijdens deze studie onderzocht, de andere stalen worden bewaard als reserve en voor stockage voor mogelijk latere analyse. U vult de dag voor de urinecollectie een vragenlijst in over de voedingswaren die u die dag hebt gegeten, en over mogelijke blootstelling aan PAK's in uw omgeving de afgelopen dag.

## Kosten

Er zijn voor u geen kosten verbonden aan dit onderzoek.

### *Risico's en ongemakken*

Er is geen enkel risico verbonden aan deze studie.

### *Voordelen*

U krijgt uw persoonlijke resultaten en de groepsresultaten van de studie.

### *Vertrouwelijkheid*

Alle informatie die u ons geeft, wordt vertrouwelijk behandeld overeenkomstig de wet op de privacy. Indien de resultaten van deze studie gepubliceerd worden in een rapport of wetenschappelijk tijdschrift zal uw naam op geen enkele manier genoemd worden.

### *Vrijwillige deelname / intrekking van deelname uit de studie*

U neemt vrijwillig deel aan de studie. U kunt op eender welk moment beslissen uw deelname aan de studie stop te zetten om gelijk welke reden, ook al ondertekende u dit toestemmingsformulier. Wij vragen u wel onze diensten hiervan op de hoogte te stellen (adres en telefoonnummers staan verder in deze brief).

### *Hoe gaat het nu verder?*

Als u wilt deelnemen, vult u bijgevoegd toestemmingsformulier in.

### *Meer informatie nodig?*

Voor vragen rond deze studie

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gudrun.koppen@vito.be

Dit voorstel werd voorgelegd aan de Adviescommissie Medische Ethiek van de Universiteit Antwerpen, een comité dat er moet voor zorgen dat deelnemers aan een wetenschappelijk onderzoek geen nadeel ondervinden.

## TOESTEMMINGSFORMULIER
## VOOR DEELNAME AAN DE STUDIE

**Onderzoek naar de oorzaak van de hogere gehalten aan PAK's bij inwoners van de regio's Menen en Genk Zuid**

**Ik heb de informatie over deze studie gelezen en begrepen. Ik neem vrijwillig deel aan deze studie.**

Wenst u een persoonlijk resultaat te ontvangen van de metingen? (1 bolletje kleuren)

○ ja, ik wens persoonlijke resultaten te ontvangen op mijn thuisadres. Bij adreswijziging zal ik mijn nieuwe adres doorgeven aan het onderzoeksteam.

○ neen, ik wens geen persoonlijke resultaten te ontvangen

**Naam: ...........................................................................**

**Adres: ...........................................................................**

**Telefoonnummer/GSM: ...................................................**

**e-mail adres: ................................................................**

**geslacht:** ○ **man** ○ **vrouw**

**geboortedatum: ..../....../19....**

**Handtekening** .......|........|........ **Datum**

## ANNEX B - GENERAL QUESTIONNAIRE

**Vragenlijst bij het project 'Inschatting bronnen van blootstelling aan PAK's**
**In te vullen bij aanvang van de studie**

Mevrouw, Mijnheer,

Met deze vragenlijst willen we informatie verzamelen over gezondheid, socio-economische gegevens, woonplaats, hobby's, verplaatsingen in verkeer, aard van verwarming, kookvuur, en verluchting. Deze informatie helpt om de meetresultaten in uw urine beter te begrijpen en te verklaren.

De vragenlijst krijgt een codenummer en uw antwoorden worden **anoniem** verwerkt. Uw naam zal in geen enkel rapport of publicatie vermeld worden. Als sommige onderdelen moeilijk of onduidelijk zijn kan u ons bellen of mailen:

| | |
|---|---|
| Els Van de Mieroop: | 03/259 12 61<br>els.vandemieroop@provincieantwerpen.be |
| Gudrun Koppen: | 014/33 51 65<br>gudrun.koppen@vito.be |

VEEL SUCCES MET HET INVULLEN VAN DE VRAGENLIJST

HARTELIJK DANK VOOR UW MEDEWERKING

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# Vragenlijst: Inschatting bronnen van blootstelling aan PAK's

Identificatienummer:

Datum bij invullen vragenlijst:………/………/……….

## A. Info over uzelf

1. Geboortedatum: ………/………/……….

2. Geslacht: **○** man **○** vrouw

3. Gewicht:……………..kg

4. Lengte:………………cm

5. Bent u alleenstaande? ○ neen
   ○ ja

6. Wat is de hoogste school- of beroepsopleiding die u en (indien van toepassing) uw partner voltooiden? Kruis aan in de tabel:

| **Diploma** | **Uzelf** | **Uw partner** |
|---|---|---|
| Geen diploma | ○ | ○ |
| Lager onderwijs | ○ | ○ |
| Lager secundair onderwijs (lager ASO, TSO, KSO of BSO) | ○ | ○ |
| Hoger secundair onderwijs (ASO, TSO, KSO, BSO) | ○ | ○ |
| Bachelor (HOBU, universiteit) | ○ | ○ |
| Master (HOBU, universiteit) | ○ | ○ |

*NB: HOBU=hoger onderwijs buiten universiteit*

7. Zijn uw biologische ouders in België geboren?
   ○ ja
   ○ neen, geboorteland moeder:……………………………
   geboorteland vader:………………………………

## B. Werk/beroep

8. Werkt u momenteel? o neen (ga direct naar deel C)
o ja

9. Op welk niveau werkt u?
   - ○ master (hoger onderwijs lange type & universiteit)
   - ○ bachelor (hoger onderwijs korte type)
   - ○ middelbaar onderwijs
   - ○ lager onderwijs
   - ○ geen vereiste scholingsgraad

10. Belangrijkste activiteit op het werk:

……………………………………………………………………………………………………………………………

……………………………………………………………………………………………………………………………

11. Postcode en naam van de gemeente/stad waar u werkt:.............................................

12. Hoeveel dagen per week bent u op de werkplek?..................................dagen/week

13. Ik ben tijdens mijn werktijd (duidt bolletjes aan):

| | Binnen | Buiten <u>zonder</u> verkeer | Buiten met <u>weinig/matig</u> verkeer | Buiten met <u>druk</u> verkeer |
|---|---|---|---|---|
| Mééste van mijn werktijd | ☐ | ☐ | ☐ | ☐ |

14. Hoe gaat u naar het werk (meerdere antwoorden mogelijk)?
   - ○ met de auto
   - ○ met de trein, tram/metro
   - ○ met de bus
   - ○ met de bromfiets/motorfiets
   - ○ met de fiets
   - ○ te voet

15. Hoe lang bent u onderweg naar het werk?....................................minuten

16. Verplaatst u zich in druk verkeer:

op weg naar het werk? o neen
o ja

op weg naar huis? o neen
o ja

## C. Hobby's

17. Hoe vaak komt u in uw vrije tijd, tijdens uw hobby of bij het klussen in contact met:

| | dagelijks | wekelijks | maandelijks | zelden | nooit |
|---|---|---|---|---|---|
| Roet, assen | ☐ | ☐ | ☐ | ☐ | ☐ |
| Uitlaatgassen* | ☐ | ☐ | ☐ | ☐ | ☐ |

**bv. in garage, tijdens motorcross, karting, quad*

18. Verplaatst u zich vaak in druk verkeer tijdens een niet-werkdag?
    - ○ neen
    - ○ ja

19. Beoefent u regelmatig sport in open lucht?
    - ○ dagelijks
    - ○ wekelijks
    - ○ maandelijks
    - ○ zelden

## D. Woning

20. Hoe oud is uw woning?.......................jaar

21. Hoeveel jaar woont u op het huidige adres?.....................jaar

22. Welk type woning hebt u?
    - ○ alleenstaande woning
    - ○ half open bebouwing
    - ○ rijwoning

23. Wat is de bewoonbare oppervlakte van uw huis?..........................................$m^2$

24. Welk aandeel van het huis is bedekt met tapijt? circa..................................$m^2$

25. Is het dak geïsoleerd?
    - ○ neen
    - ○ ja

26. Is er een open verbinding tussen uw keuken en leefruimte?
    - ○ neen
    - ○ ja

27. Hoe verwarmt u uw woning? (u mag meer dan één mogelijkheid aankruisen)
    - ○ Gas
    - ○ Mazout
    - ○ Elektriciteit
    - ○ Andere : ……………………………..

28. Welke energie gebruikt u om te koken?
    - ○ Gasfornuis
    - ○ Elektrische kookplaat
    - ○ Andere : ……………………………..

29. Hoeveel mensen wonen er in uw woning?

    Aantal volwassenen: ..........
    Aantal kinderen: ..........

30. Hoeveel dagen per week bent u op dit adres: …………….dagen/week

31. Welk type garage heeft u?
    - ○ geen garage
    - ○ een garage voor 2 wagens met een deur naar de keuken
    - ○ een garage voor 2 wagens met een deur naar een andere kamer
    - ○ een garage voor 1 wagen met een deur naar de keuken
    - ○ een garage voor 1 wagen met een deur naar een andere kamer
    - ○ een losstaande garage

32. Bestaat uw oprit uit asfalt?
    - ○ neen, niet uit asfalt of heb geen oprit (ga naar vraag 32)
    - ○ ja

….

33. Indien uw oprit bestaat uit asfalt, heeft u deze het afgelopen jaar nog behandeld met bitumen?
    - ○ neen
    - ○ ja

34. Gebruikt u mottenballen/ deo blokken in de kleerkast?
    - ○ neen
    - ○ ja

35. Zijn er soms vreemde geuren in uw huis?
    - ○ Geen vreemde geuren
    - ○ Verfgeur
    - ○ Benzinegeur
    - ○ Geur van verbranding
    - ○ Andere:……………………………….

36. Zijn er in de buurt bepaalde verontreinigingen/activiteiten die mogelijk een verhoogde blootstelling aan **verbrandingsproducten, teer, uitlaatgassen**,… kunnen veroorzaken (stoken, verkeer, asfaltwerken,…)?

……………………………………………………………………………………………………………………………

……………………………………………………………………………………………………………………………

**E. Roken/passief roken**

37. Hebt u ooit gerookt?
    - ○ neen, nooit gerookt → ga naar vraag 38
    - ○ ja

38. Rookt u momenteel nog?
    - ○ neen
    - ○ ja

39. Hoe lang heeft u in totaal gerookt ? *(tel al de periodes op)*

☐☐ weken ☐☐ maanden ☐☐ jaren

40. Hoeveel uren per week bent u gemiddeld in de aanwezigheid van mensen die roken (week + weekend samengeteld)?

☐☐ uren per week

**Hartelijk dank voor het invullen van de vragenlijst!**

## ANNEX C – INSTRUCTIONS FOR COLLECTION OF URINE

### INSTRUCTIES URINE VERZAMELEN

De verzameling van de urinestalen gebeurt gedurende 4 weken. In elke week van deze 4 weken, dient volgende te worden uitgevoerd:

- Op **woensdagavond** vult u de voedingsvragenlijst in over wat je de afgelopen dag hebt gegeten.
- Op **donderdag** van elke week vangt u een deel van de ochtendurine op in het urinepotje dat u gekregen hebt. *OPMERKING: Indien u die dag vergeet de urine te verzamelen, kan u ook nog urine opvangen op de zaterdag die erop volgt. In dat laatste geval vul je een voedingsvragenlijst in op vrijdagavond.*
- De urinepotjes zijn genummerd.
- Haal het potje uit het plastic zakje, controleer of de datum juist is en noteer het uur op het invulformulier onderaan.
- Open de schroefdop en plas rechtstreeks in het urinepotje. Let op: niet volledig vol maken. Indien dit het geval zou zijn, kan je een beetje uitgieten in het toilet.
- Schroefdop terug op het potje plaatsten en het geheel in het plastic zakje steken.
- Bewaar dit potje in de diepvriezer (-20°C) en vul de bijhorende urinevragenlijst in.

**Identificatienummer deelnemer**:…………………………………………………………………………….

*Invulformulier voor inventarisatie van de gevulde urinepotjes*

| Week | Identificatienummer urinepotje | Datum | Uur | Opmerkingen |
|---|---|---|---|---|
| **1** | Urine 1 | | | |
| **2** | Urine 2 | | | |
| **3** | Urine 3 | | | |
| **4** | Urine 4 | | | |

## ANNEX D – QUESTIONNAIRE ACCOMPANIED WITH URINE COLLECTION

# Vragenlijst bij urine-afname

Identificatienummer:

Datum van vandaag: ………/………./ ……………

## A. Gezondheid op dit moment

1. Bent u de laatste week ziek geweest?
   - ○ neen
   - ○ ja

2. Neemt u nu medicatie?
   - ○ neen
   - ○ ja, welke?..........................

## B. Blootstelling de dag voor de urinecollectie

3. Kwam u de laatste 24 uur langdurig in contact met uitlaatgassen, bv. in een garage, bij motorcross, karting, quads (de normale verkeersblootstelling niet meegerekend)?
   - ○ neen
   - ○ ja

4. Heeft u de laatste 24 uur binnenshuis etherische olie gebruikt?
   - ○ neen
   - ○ ja

5. Heeft u de laatste 24 uur binnenshuis olielampjes, kaarsen, geurkaarsen of wierookstokjes gebruikt?
   - ○ neen
   - ○ ja

6. Hoeveel tijd hebt u de laatste 24 uur in een rokerige omgeving doorgebracht?

   ☐☐ uren

7. Had u gisteren bezoekers, die in uw huis hebben gerookt?
   - ○ neen
   - ○ ja

8. Heeft het de laatste 24 uur geregend?
   - ○ neen
   - ○ ja

9. Heeft u de laatste 24 uur een raam geopend/verlucht?
   - ○ neen
   - ○ ja

10. hebt u deze nacht met open venster geslapen?
    - ○ neen
    - ○ ja

11. Heeft u gisteren avond thuis gekookt?
    - ○ neen
    - ○ ja

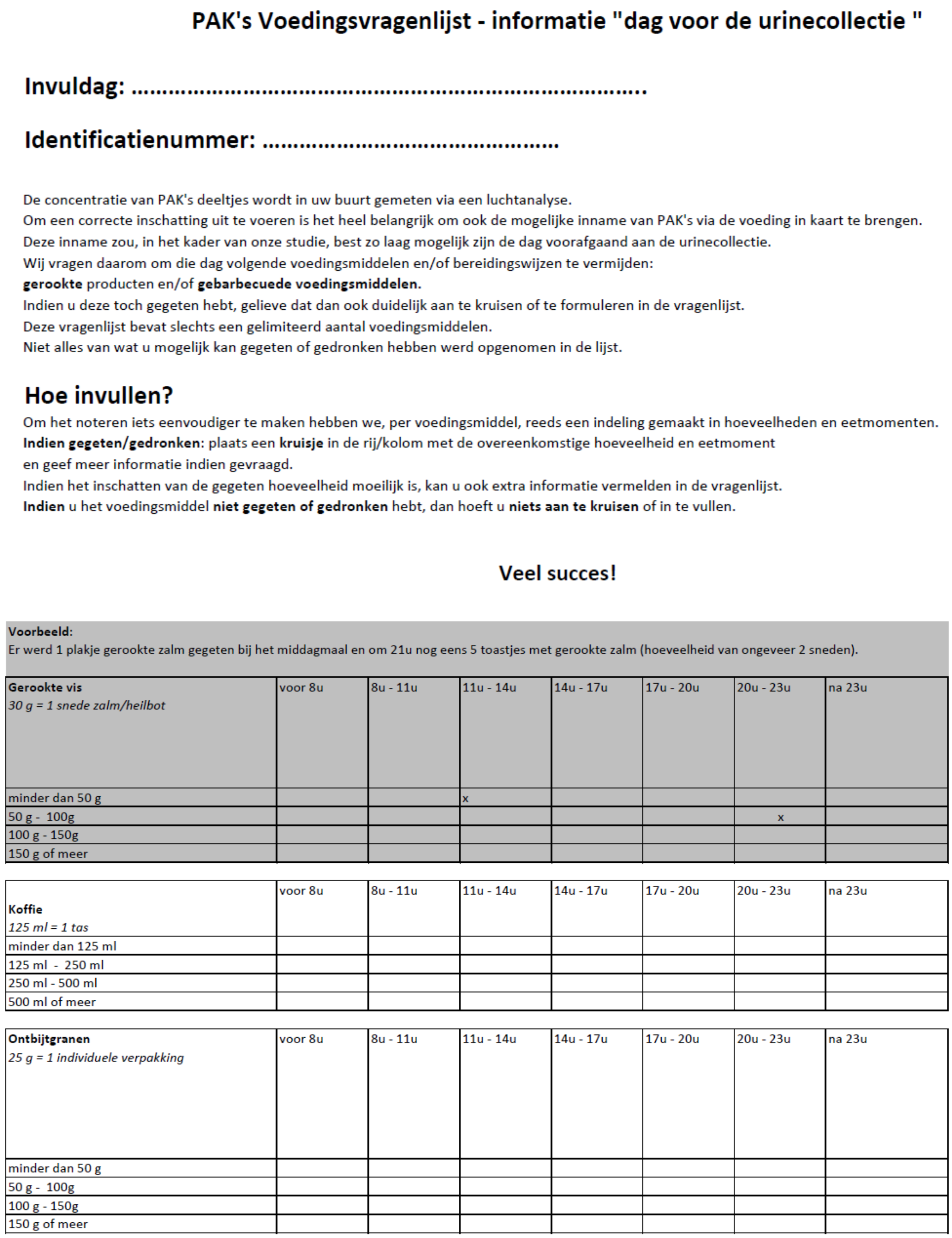
12. Heeft u de laatste 24 uur de verwarming aangezet?
    - ○ neen
    - ○ ja

13. Heeft u de laatste 24 uur een vreemde geur (benzine, rook) waargenomen?
    - ○ neen
    - ○ ja, de geur was <u>binnenshuis</u> waar te nemen
    - ○ ja, de geur was <u>buitenshuis</u> waar te nemen

14. Hebt u de laatste 24 uur buitenshuis gestookt?
    - ○ neen
    - ○ ja

**Hartelijk dank voor het invullen van de vragenlijst!**

## ANNEX E – FOOD QUESTIONNAIRE



### PAK's Voedingsvragenlijst - informatie "dag voor de urinecollectie "

**Invuldag: ……………………………………………………………………..**

**Identificatienummer: ……………………………………….**

De concentratie van PAK's deeltjes wordt in uw buurt gemeten via een luchtanalyse.
Om een correcte inschatting uit te voeren is het heel belangrijk om ook de mogelijke inname van PAK's via de voeding in kaart te brengen.
Deze inname zou, in het kader van onze studie, best zo laag mogelijk zijn de dag voorafgaand aan de urinecollectie.
Wij vragen daarom om die dag volgende voedingsmiddelen en/of bereidingswijzen te vermijden:
**gerookte** producten en/of **gebarbecuede voedingsmiddelen.**
Indien u deze toch gegeten hebt, gelieve dat dan ook duidelijk aan te kruisen of te formuleren in de vragenlijst.
Deze vragenlijst bevat slechts een gelimiteerd aantal voedingsmiddelen.
Niet alles van wat u mogelijk kan gegeten of gedronken hebben werd opgenomen in de lijst.

### Hoe invullen?

Om het noteren iets eenvoudiger te maken hebben we, per voedingsmiddel, reeds een indeling gemaakt in hoeveelheden en eetmomenten.
**Indien gegeten/gedronken**: plaats een **kruisje** in de rij/kolom met de overeenkomstige hoeveelheid en eetmoment
en geef meer informatie indien gevraagd.
Indien het inschatten van de gegeten hoeveelheid moeilijk is, kan u ook extra informatie vermelden in de vragenlijst.
**Indien** u het voedingsmiddel **niet gegeten of gedronken** hebt, dan hoeft u **niets aan te kruisen** of in te vullen.

**Veel succes!**

**Voorbeeld:**
Er werd 1 plakje gerookte zalm gegeten bij het middagmaal en om 21u nog eens 5 toastjes met gerookte zalm (hoeveelheid van ongeveer 2 sneden).

| **Gerookte vis**<br>*30 g = 1 snede zalm/heilbot* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | x | | | | |
| 50 g - 100g | | | | | | x | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Koffie**<br>*125 ml = 1 tas* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 125 ml | | | | | | | |
| 125 ml - 250 ml | | | | | | | |
| 250 ml - 500 ml | | | | | | | |
| 500 ml of meer | | | | | | | |

| **Ontbijtgranen**<br>*25 g = 1 individuele verpakking* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Muesli/cruesli**<br>*10 g = 1 eetlepel* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Brood**<br>*30 g = gemiddelde snede* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Pistolet, sandwich , stokbrood**<br>*40 g = 1 pistolet/sandwich/piccolo* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Gerookte vis**<br>*30 g = 1 snede zalm/heilbot* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Gerookte vleeswaren:**<br>*50 g = 1 snede hesp/4 sn. bacon of spek* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Gerookte kaas, Haloumi**<br>*25 g = 1 individuele verpakking kaas* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Chips, gezouten versnaperingen**<br>*45 g = 1 klein zakje chips* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 50 g | | | | | | | |
| 50 g - 100g | | | | | | | |
| 100 g - 150g | | | | | | | |
| 150 g of meer | | | | | | | |

| **Noten**<br>*25 g = 1 handje / 20g = 1 eetlepel* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 25 g | | | | | | | |
| 25 g - 50 g | | | | | | | |
| 50 g - 75 g | | | | | | | |
| 75 g of meer | | | | | | | |

| **Mayonaise, dressing, slasaus**<br>*25 g = 1 eetlepel mayonaise*<br>*10g = 1 eetlepel vinaigrette* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 25 g | | | | | | | |
| 25 g - 50 g | | | | | | | |
| 50 g - 75 g | | | | | | | |
| 75 g of meer | | | | | | | |

| **Olie om te bakken en braden**<br>*10 g = 1 eetlepel* | voor 8u | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u |
|---|---|---|---|---|---|---|---|
| minder dan 20 g | | | | | | | |
| 20 g - 40 g | | | | | | | |
| 40 g - 60 g | | | | | | | |
| 60 g of meer | | | | | | | |

Welke soort olie(n)? ………………………………………………………………

**Hebt u de dag voor de urinecollectie aangebrande voedingsmiddelen gegeten?**

○ neen
○ ja, niet gebarbecued
○ ja, wel gebarbecued

**Hebt u de dag voor de urinecollectie gebarbecuede voedingsmiddelen gegeten?**

○ neen
○ ja: gelieve in de tabel hieronder te noteren wat én hoeveel hiervan u juist gegeten hebt en op welk eetmoment.
Druk de hoeveelheden uit in huishoudelijke maten zoals sneden, stuks, eetlepels, …. Vermijd vage hoeveelheden zoals dun, veel, ….

**Voorbeeld:**

| **Wat?** | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u | direct contact met de vlammen: |
|---|---|---|---|---|---|---|---|
| *varkenskotelet, filet: 1 stuk* | | | x | | | | ● ja ○ neen ○ ? |
| *merguez worst: stukje van 5 cm* | | | x | | | | ○ ja ● neen ○ ? |
| *aubergine: 3 dunne schijfjes, niet geschild* | | | x | | | | ○ ja ● neen ○ ? |

| **Wat?** | 8u - 11u | 11u - 14u | 14u - 17u | 17u - 20u | 20u - 23u | na 23u | direct contact met de vlammen: |
|---|---|---|---|---|---|---|---|
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |
| | | | | | | | ○ ja ○ neen ○ ? |

**Welke vorm van hittebron werd er gebruikt?**

○ houtskool ○ houtskool/houtsnippers ○ briketten ○ gas

**Hartelijk bedankt voor uw deelname!**

## ANNEX F – INSTRUCTIONS FOR AIR SAMPLING BY PARTICIPANTS

# INSTRUCTIES voor verzameling van de LUCHTSTALEN

## 1. WERKWIJZE PATRONEN VERVANGEN voor verzamelen van PAK's

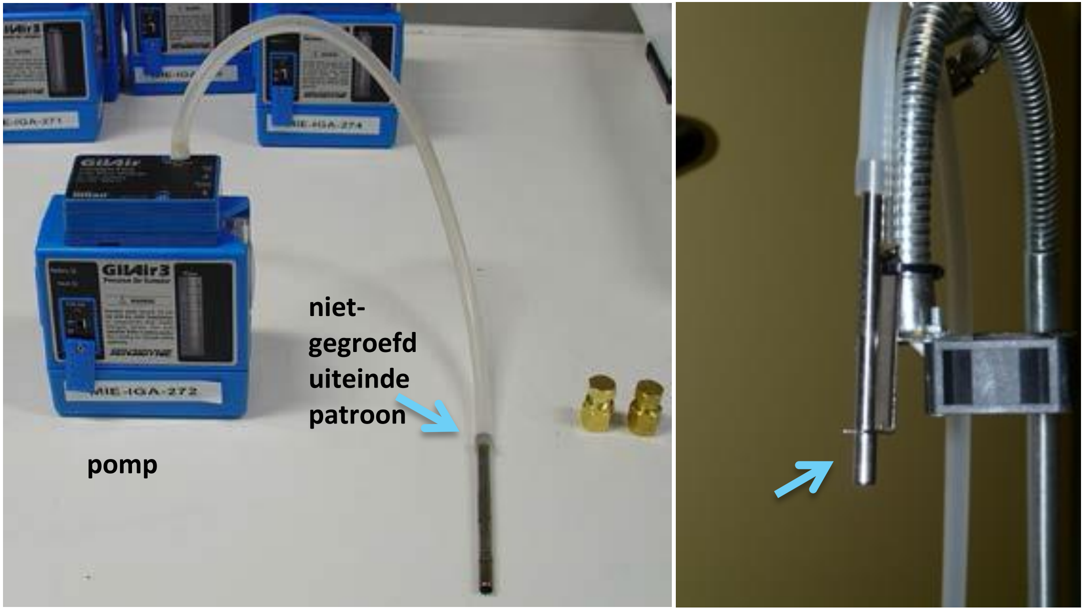
### *Bewaring patronen voor en na gebruik*

Alle patronen (patroon 1, 2, 3…8) bewaren op kamertemperatuur gewikkeld in aluminiumfolie, in een plastiek zakje. Zeker **NIET** koel bewaren!

### *Start bemonstering*

1. Bij elke start van monstername moet een nieuw patroon genomen worden.
2. Koperkleurige afdichting aan beide kanten van het adsorptiepatroon losmaken met de meegeleverde aluminium sleutels (sleutelsysteem, zie Figuur 1). De koperkleurige afdichtingen zijn geen wegwerpmateriaal en dienen bewaard te worden om de adsorptiepatronen na de bemonstering terug af te sluiten!
3. Darm van de pomp aan het niet-gegroefde uiteinde aankoppelen. Klik het gegroefde uiteinde van het patroon neerwaarts vast in de houder op de staander (zie figuur2).
4. Pomp aanzetten
5. Opschrijven van startdatum/uur/minuut op het invulformulier (zie tabel helemaal onderaan).

*Figuur 1: patroon met koperkleurige afdichtingen aan beide uiteinden. Deze afdichtingen worden geopend met aluminium sleutelsysteem (zie op de foto aangebracht op linkse uiteinde van het patroon)*



*Figuur 2: Montage van patroon: patroon aan het niet-gegroefde uiteinde aan de darm met de pomp koppelen (zie pijltje op de linkse foto). Het gegroefde uiteinde (onderaan) vastklikken in de houder (zie pijltje op de rechtse foto).*

## *Stop bemonstering*

- Na 2 dagen, dit is ca. 48 uur na de start, pompje uitzetten.
- Adsorptiepatroon loskoppelen van de staander en terug dichtschroeven aan beide uiteinden met de meegegeven sleutels.
- Opschrijven van einddatum/uur/minuut op invulformulier.
- Alle onderdelen bewaren op kamertemperatuur (**zeker NIET koelen!!**). De veldwerkers van VITO of PIH nemen contact met u op om een afspraak te maken voor het ophalen van de stalen.
- Opnieuw starten van volgende bemonstering (zie start bemonstering op pagina 1).

## 2. WERKWIJZE FILTERS VERVANGEN voor staalname van levoglucosan in stof met Harvard impactor en pomp

### *Bewaring filters voor en na gebruik*

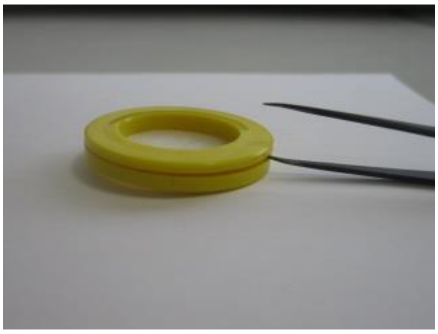
Alle filters zitten in een afzonderlijke plastieken behuizing (petriplaat) met daarop de nummer (filter 1, 2,…..8). De filters worden voor en na gebruik KOEL bewaard in de koelkast en in de passende behuizing.

### *Start bemonstering*

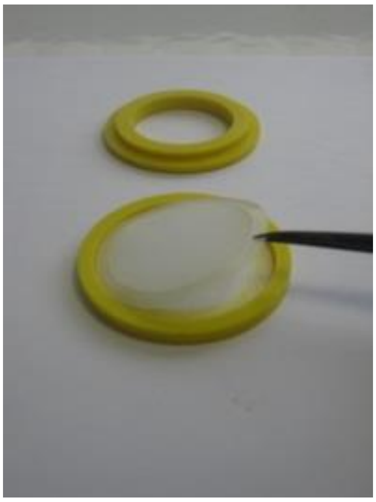
1. Bij elke start van bemonstering moet een nieuwe filter geplaatst worden.
2. Open de Harvard impactor, door onderaan aan de zijkant beide kleppen te openen:

→

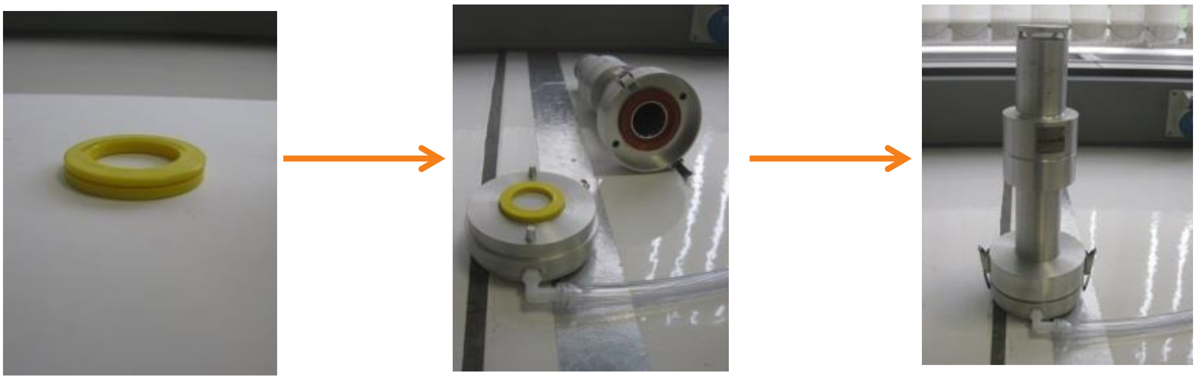
3. Neem de gele filtercasette uit de boven afgebeelde impactor en open de gele casette met behulp van het pincet, zoals in foto hier onder afgebeeld:



4. Neem een nieuwe filter uit de plastieken petriplaat met behulp van het pincet. Sluit de petriplaat terug. De nieuwe filter met behulp van het pincet in de gele filtercasette (op het steungaas) monteren:



5. Sluit de gele filtercasette waarin je de filter hebt geplaatst en plaats deze casette in de impactor. Sluit de impactor met beide klemmen aan de zijkant:



6. Pomp aanzetten. Afhankelijk van het pomptype krijg je bij het opstellen van de pomp, een handleiding en instructies hoe de pomp aan en uit te zetten.

   pomp type Tecora : zet de pomp aan m.b.v. de contactsleutel
   pomp type KNF Neuberger N035AN.18 : zet de pomp aan m.b.v. de aan/uit schakelaar
   monsternamekast Vito : zet de pomp aan m.b.v. de aan/uit schakelaar

7. Opschrijven van startdatum/uur/minuut op het invulformulier (zie tabel helemaal onderaan).

## *Stop bemonstering*

1. Na 2 dagen of ca. 48 uur na de start, de pomp afzetten.

2. Open onderaan beide klemmen van de impactor en neem de gele filtercasette uit de impactor.

3. Filter met behulp van het pincet uit de gele filtercasette nemen en in de juiste petriplaat leggen. L**et op: het steungaas in de filtercasette laten liggen!**

4. De petriplaat verder in de koelkast bewaren (2-8°C). De veldwerkers van VITO of PIH nemen contact met u op om een afspraak te maken voor het ophalen van de stalen.

5. Een nieuwe, volgende filter met behulp van het pincet in de gele filtercasette plaatsen (zoals hierboven beschreven).

6. Opnieuw starten van de volgende bemonstering (zie: start bemonstering op pagina 3).

*Tabel Invulformulier voor noteren van datum/uur opstarten en afzetten van de lucht-staalname-apparatuur*

**Identificatienummer deelnemer:……………….………………………………………………**

| WEEK | Identificatie(ID) | START *aankoppelen van patroon of plaatsen filter en opzetten van pomp* | | STOP *afzetten pomp en loskoppelen van patroon of verwijderen filter* | | Opmerkingen |
|---|---|---|---|---|---|---|
| | | datum | Uur:min (vb 10:30) | datum | Uur:min (vb 10:45) | |
| week1 31 aug – 6 sept | **patroon1:**<br>**filter 1:** | | | | | |
| | **patroon 2:**<br>**filter 2:** | | | | | |
| week2 7-13 sept | **patroon 3:**<br>**filter 3:** | | | | | |
| | **patroon 4:**<br>**filter 4:** | | | | | |
| Week 3: 14-20 sept. | **patroon 5:**<br>**filter 5:** | | | | | |
| | **patroon 6:**<br>**filter 6:** | | | | | |
| Week 4: 21-27 sept. | **patroon 7:**<br>**filter 7:** | | | | | |
| | **patroon 8:**<br>**filter 8:** | | | | | |

## ANNEX G - SCHEME FOR SAMPLE COLLECTION BY PARTICIPANTS

### DAGSCHEMA staalname PAK's studie

Het schema voor verzameling van luchtstalen, urine en invullen van de vragenlijsten wordt schematisch weergegeven in de tabel onderaan. De collectie van luchtstalen gebeurt aan de gevel, in de voortuin of achtertuin van je huis gedurende 4 weken.

In elke week van deze 4 weken, dient volgende worden uitgevoerd:

- Twee **lucht-samplers** opzetten, beide op dinsdag en donderdag (in de tabel aangeduid met 'patroon + filter').
- Een **Vragenlijst** over blootstelling aan PAK's de laatste 24u, invullen op woensdag van elke week.
- Urine verzamelen op donderdagochtend bij het opstaan.

*Schema van de taken die worden uitgevoerd in de 4-weken staalnameperiode*

| Dag | Week 1 | Week 2 | Week 3 | Week 4 |
|---|---|---|---|---|
| Maandag | Installatie luchtsamplers + afleveren materiaal door veldwerkers | | | |
| Dinsdag | Patroon+filter 1 opstarten | Patroon+filter 3 opstarten | Patroon+filter 5 opstarten | Patroon+filter 7 opstarten |
| Woensdag | **Vragenlijst 1** | **Vragenlijst 3** | **Vragenlijst 5** | **Vragenlijst 7** |
| Donderdag | **Ochtendurine 1**<br>Patroon+filter 1 verwijderen en 2 opstarten | **Ochtendurine 2**<br>Patroon+filter: 3 verwijderen en 4 opstarten | **Ochtendurine 3**<br>Patroon+filter 5 verwijderen en 6 opstarten | **Ochtendurine 3**<br>Patroon+filter 7 verwijderen en 8 opstarten |
| Vrijdag | | | | |
| Zaterdag | Patroon+filter 2 verwijderen | Patroon+filter 4 verwijderen | Patroon+filter 6 verwijderen | Patroon+filter 8 verwijderen |
| zondag | | | | |