AIR POLLUTION FROM RESIDENTIAL WOOD COMBUSTION IN A DANISH VILLAGE

Measuring campaign and analysis of results

NERI Technical Report no. 777 2010

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Data sheet

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Abstract: A campaign took place in the winter 2006/2007 comprising measurements of many air pollution components at two sites: a wood smoke exposed site within the village Slagslunde, and a background site 500 m outside of the village. The report describes the campaign and its results. A central result is a so-called 'wood smoke source profile', which relates several measures of wood smoke pollution to each other. This is based on a ‘cleaned’ data set, for which the effect of other sources than wood smoke was small. The wood smoke profile links the measures $PM_{2.5}$, particle volume, soot, monosaccharide anhydrides (levoglucosan and mannosan) and different PAHs to each other. Particle number N does not have a close link to the other measures.

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Baggrund

Forskningsprojektet WOODUSE er et tværfagligt projekt, der dækker mange aspekter omkring brændefyring. Det omfatter bl.a. undersøgelser af emissioner, forureningsniveauer i udeluft og indeluft, helbredseffekter samt sociale aspekter. Projektet har haft deltagelse af 4 institutioner (Danmarks Miljøundersøgelser ved Aarhus Universitet, Det Nationale Forskningscenter for Arbejdsmiljø, Institut for Folkesundhedsvidenskab ved Københavns Universitet samt Institut for Byggeri og Anlæg ved Danmark Tekniske Universitet). Projektet er støttet af Det Strategiske Forskningsråd (bevilling ENMI-2104-05-0010, perioden 2006-2009).

Som en del af WOODUSE foregik en tre måneders målekampagne i Slagslunde i vinteren 2006-2007 med henblik på at bestemme brændefyrings bidrag til den lokale luftforurening. Nærværende rapport beskriver målekampagnen og dens resultater. Rapporten har en relativt teknisk karakter, men den suppleres af en lettere tilgængelig dansksproget rapport (Olesen et al., 2010b), som gengiver centrale resultater og bygger videre på dem ved hjælp af modelberegninger.

Formålet med målekampagnen var at vurdere betydningen af den lokale brændefyring på luftkvaliteten. Det var derfor vigtigt at identificere andre mulige kilder og at kvantificere eller eliminere deres bidrag. Slagslunde blev udvalgt som egnet målested, fordi det er en lille by med velafgrænset udstrækning, og fordi der er få andre kilder til luftforurening end fyring med brændeovne. De vigtigste andre lokale kilder er trafik og et mindre kraftvarmeværk, der bruger naturgas. Værket har en 20 meter høj skorsten og ligger på den lokale folkeskoles område i den vestlige del af landsbyen. Der er en begrænset trafik på lokale biveje nær ved målestederne. Det var forventet, at effekten af disse kilder ville være lille.

Alle måleinstrumenter blev anbragt i to mobile målestationer. Den ene tæt på huse, hvor der fyres med brændeovne i midten af landsbyens sydlige halvdel, og den anden som baggrundsreference 500 meter VNV for landsbyen. Ved at opstille to målestationer tæt på hinanden er det muligt at eliminere indflydelsen fra langtransport samt regionale kilder, idet man betragter forskellen mellem koncentrationerne målt inde i Slagslunde og koncentrationerne målt i baggrunden udenfor landsbyen. Placeringen af baggrundsstationen er mere optimal end i tidligere studier, hvor baggrundsmålingerne blev foretaget i en afstand på mange kilometer.

Hovedresultater

Et væsentligt resultat fra målekampagnen er en såkaldt "kildeprofil for brænderøg", som relaterer forskellige målestokke for forurening med brænderøg til hinanden. Forurening med brænderøg viser sig ved øget koncentration af fine partikler ($PM_{2.5}$), men også ved øget koncentration af sod, ved øget partikelvolumen, samt på andre måder. Kildeprofilen er et nyttigt værktøj i forbindelse med analyser af fremtidige målinger.

Brænderøgs bidrag til niveauet af $PM_{2.5}$ for målevognen i Slagslunde blev bestemt til 2,0 µg/m^3 som gennemsnit for en periode på ca. ni uger i en mild vinter.

Rapporten demonstrerer hvorledes målinger af størrelsesfordelingen af partikler er nyttige til at identificere forskellige kilders bidrag, og der gøres rede for brænderøgs indflydelse på størrelsesfordelingen af partikler.

Der blev foretaget målinger af PAH (tjærestoffer) i et beskedent antal dage (10 døgn). Målingerne viste at PAH-niveauet i brændeovnskvarteret var kraftigt forhøjet i forhold til baggrundsniveauet. Datagrundlaget er for lille til at drage nogen konklusion om niveauet af PAH på årsbasis.

Levoglucosan og mannosan benyttes i andre undersøgelser som indikatorer for brænderøg. Det fremgår imidlertid af arbejdet med etablering af en kildeprofil, at målinger af disse stoffer ikke egner sig til at kvantificere brændefyrings bidrag til $PM_{2.5}$.

Målekampagnen

Slagslunde er en mindre landsbybebyggelse med ca. 400 parcelhuse, som næsten alle er i et plan. Landsbyen har karakter af soveby uden lokal industri og er omgivet af åbent land med landbrug. Ca. halvdelen af husene er udstyret med brændeovne.

Der blev gennemført målinger fra en målevogn centralt i Slagslunde og fra en baggrundsstation ca. 500 meter uden for byen. Samtidige målinger med alle måleinstrumenter foregik i fyringssæsonen i perioden fra 23. december 2006 til 26. februar 2007. Forbruget af træ til fyring i de enkelte huse blev bestemt på grundlag af spørgeskemaer, som beboerne udfyldte.

Landsbyen ligger på en aflang bakke. De meteorologiske målinger blev foretaget på en mast placeret på bakkens top 500 meter NV for landsbyen.

Under den eksperimentelle del af kampagnen blev der foretaget kontinuerte målinger af en række luftforureningskomponenter: $PM_{2.5}$, partikelantal, størrelsesfordeling af partikelantal, CO, NO og NO_x, samt sodsværtning.

Udover de kontinuerte målinger blev der foretaget 10 døgnopsamlinger af partikler og semiflygtige forbindelser til kemisk analyse for PAH, levoglucosan og mannosan. PAH er kræftfremkaldende, mens levoglucosan og mannosan er interessante, fordi de er karakteristiske for afbrænding af biomasse.

I en mindre, to uger lang kampagne blev forholdet mellem indendørs- og udendørs luftforurening undersøgt i to huse: et med brændeovn og et uden.

Behandling af data

Analysen af måleresultaterne tog i høj grad sigte på at identificere andre mulige kilder end brændefyring.

Formålet med målingerne af NO_x (og NO) var at kunne foretage korrektioner for trafikken, som er hovedkilde til NO_x, men som også i mindre grad kan bidrage til partikelantallet (N) og til $PM_{2.5}$. Fra andre undersøgelser er N/NO_x-forholdet og $PM_{2.5}/NO_x$-forholdet kendt for trafikemissioner. Målinger af NO_x kan derfor bruges til at beregne bidragene fra trafikken. Kvalitetskontrollen af NO_x-data efter kampagnen viste uheldigvis, at ingen af de to NO_x-monitorer havde fungeret tilfredsstillende i hele kampagneperioden, og at der kun var 23 dage med godkendte samtidige målinger. Målingerne kunne derfor ikke bruges til at korrigere for trafikbidraget. På grundlag af målingerne i de 23 dage blev det vurderet, at en lille men ikke helt ubetydelig del af det lokale bidrag til partikelantallet (10-20 %) kan skyldes den lokale trafik. For $PM_{2.5}$ kunne det imidlertid vises, at det lokale trafikbidrag var ubetydeligt (< 5 %).

Målingerne af størrelsesfordelingen af partikelantallet, beregnet som fordelingen inde i Slagslunde minus fordelingen på baggrundsstationen (inkrementet), viste sig at være velegnede til at afsløre andre mulige forstyrrende kilder. En top af meget små partikler omkring 13 nm var undertiden til stede i størrelsesfordelingen, men kun i perioder med bestemte vindretninger. Toppen var positiv ved vindretninger omkring 280° og negativ ved vindretninger omkring 140°. Dette er præcis retningerne til det lokale kraftvarmeværk fra henholdsvis målestedet i Slagslunde og baggrundsmålestedet uden for Slagslunde. Ved andre vindretninger, eller når gasturbinen på kraftvarmeværket var slukket sent om natten, var der ingen 13 nm top. Episoder med tydelige bidrag fra lokal trafik kunne på lignende måde identificeres ved hjælp af en top i partikelspekteret ved 26 nm.

På grundlag af målingerne af partikelstørrelsesfordelinger var det muligt at konstruere et 'renset' datasæt, for hvilket det kan antages, at bidragene til inkrementerne fra andre kilder end lokal fyring med brændeovne er minimal. Data målt under episoder med tydelige bidrag fra trafik, samt data målt i vindretningsintervaller omkring de to ovennævnte retninger til kraftvarmeværket, blev udeladt i det rensede datasæt. Dette datasæt, som udgør 62 % af det samlede datasæt, blev brugt til den videre analyse. Hvorvidt man brugte hele datasættet, eller kun det rensede datasæt, betød imidlertid kun en signifikant forskel for partikel*antallet*. Årsagen er, at både trafikken og kraftvarmeværket producerer forholdsvis mange partikler, men meget lidt masse, partikelvolumen og sod i sammenligning med brændeovnene.

Døgnvariationen (beregnet på grundlag af timemiddelværdier for hele det rensede datasæt), viser forhøjede koncentrationer fra midt på eftermiddagen og om aftenen, hvilket svarer godt til fyringsaktiviteten i Slagslunde, som blev detaljeret opgjort ved spørgeskemaundersøgelsen.

Da døgnvariationen af $PM_{2.5}$, partikelvolumen (V) og sod er næsten ens, peger det på, at kun én væsentlig kilde (brændeovnene) er årsag til inkrementerne ved målestationen i landsbyen. Døgnvariationen af partikelantallet afviger en smule fra de andre, idet koncentrationen midt på dagen er relativt forhøjet, hvilket kan skyldes partikler fra trafik.

Kildeprofil for brænderøg

Ved hjælp af receptormodelværktøjet COPREM blev beregnet en "kildeprofil for brænderøg" med veldefinerede komponenter for $PM_{2.5}$, V og sod. Komponenten for partikelantal var mere usikker, hvilket som ovenfor nævnt peger på, at der er bidrag fra andre kilder end brændefyring.

Inkrementer for PAH'er over detektionsgrænsen blev beregnet på grundlag af syv døgnopsamlinger, som foregik samtidigt på de to målesteder, og for hvilke der var samtidigt målte inkrementer af $PM_{2.5}$, V og sod, således at kildestyrken for brændefyring bestemt med COPREM var kendt. En regressionsanalyse viste, at inkrementet for PAH udviste ret dårlig korrelation med $PM_{2.5}$, V og sod, hvilket afspejles i meget store usikkerheder på PAH-værdierne i den beregnede kildeprofil.

Man har i andre undersøgelser af forurening fra brændefyring benyttet levoglucosan og mannosan som indikatorer for brænderøg, fordi de dannes netop ved afbrænding af biomasse. Inkrementer af levoglucosan og mannosan målt ved ni døgnopsamlinger blev derfor undersøgt med brug af COPREM, og brændefyringsprofilen for forholdene mellem levoglucosan, mannosan, $PM_{2.5}$, V og sod blev beregnet. Imidlertid udviste de to stoffer ringe korrelation med $PM_{2.5}$, V og sod, hvilket afspejler sig ved store usikkerheder på levoglucosan og mannosan i profilen.

Et centralt og nyttigt resultat af undersøgelsen i Slagslunde er en veldefineret 'kildeprofil for brænderøg', d.v.s. en vektor, som forbinder de tre forureningskomponenter $PM_{2.5}$, V og sod med hinanden, således at man ved måling af bidraget fra en af dem kan estimere bidragene fra de to andre. Derimod er sammenhængen til tilsvarende bidrag for partikelantal, PAH'er, levoglucosan og mannosan i døgnprøver ikke god. De kan kun beregnes med stor usikkerhed. Undersøgelsen peger således på, at ganske vist er levoglucosan og mannosan specifikke markører for afbrænding af biomasse, men de egner sig ikke til at kvantificere, hvor stort koncentrationsbidraget af $PM_{2.5}$ er.

Den resulterende kildeprofil for brænderøg, som viser gennemsnitsbidragene til koncentrationerne for hele kampagnen (for det rensede datasæt), var:

$PM_{2.5}$	1,91±0.05	$\mu g m^{-3}$
V	1,78±0.07	$\mu m^3 cm^{-3}$
Sod	2,01±0.08	Mm^{-1} (d.v.s. 'per Megameter')

PAH

Niveauet af PAH i brændeovnskvarteret var klart forøget i forhold til baggrundsstationen. For benz(a)pyrene (BaP) var koncentrationen i gennemsnit forøget med en faktor 2,3 til 1,68 ng/m^3. Det forhåndenværende datamateriale er alt for begrænset til konklusioner vedrørende årsgennemsnittet, men til sammenligning kan nævnes, at EU's målværdi for koncentrationen af BaP på årsbasis er 1 ng/m^3.

Indendørs luftforurening

Resultaterne fra den mindre målekampagne, hvor indendørs koncentrationer blev målt i to huse, behandles ikke detaljeret her i rapporten. Dog er det værd at notere følgende iagttagelser:

- I et hus *uden* brændeovn var koncentrationen af sod væsentligt lavere end udenfor.
- I et hus *med* brændeovn var koncentrationen af sod i gennemsnit nogenlunde den samme som udenfor, men der var stor tidsvariation. Hver gang der blev fyret i en kold brændeovn, måltes en stigning til værdier langt over udendørsniveauet. Derefter faldt niveauet gradvist over 5-7 timer til omkring det samme som udenfor. Når der ikke blev fyret, var niveauet lavere end udenfor.

Summary

Background

WOODUSE is a comprehensive research project, which covers a wide range of issues related to residential wood combustion. It includes investigations of emissions, air pollution level in outdoor and indoor air, health effects and social aspects. The project has had participation by four Danish institutions (National Environmental Research Institute at Aarhus University, National Research Centre for the Working Environment, Department of Public Health, University of Copenhagen, Department of Civil Engineering, Technical University of Denmark). The project has received funding from the Strategic Research Council (grant ENMI-2104-05-0010, period 2006-2009).

In the winter 2006/7 a three month measuring campaign was conducted as part of the WOODUSE project. The campaign was targeted at studying the effect of residential wood combustion to air quality, and it took place in the village of Slagslunde in the countryside North of Copenhagen. The present report describes the campaign and its results.

The report is intended for a technically minded audience. It is supplemented by a more easily accessible report in Danish (Olesen et al., 2010b), which presents central results and extends them with the aid of model calculations.

The purpose of the campaign was to assess the effect of local residential wood combustion on air quality. Therefore, it was important to identify other potential sources and to quantify or eliminate their contribution. Slagslunde was selected as a suitable site because it is confined in extent, and there are only few sources other than wood combustion. The most important other sources are local traffic and a small combined power and district heating station using natural gas. This station has a 20 meter high stack and is positioned at the grounds of the public school in the western part of the village. The traffic near the sampling sites is modest. The effect of these other sources was expected to be small.

All air pollution monitoring instruments were placed in two mobile monitoring stations, one exposed to smoke from woodstoves at a site in the centre of the southern part of the village, and the other for background reference 500 meters outside the village to the WNW. This setup makes it possible to eliminate the influence from long range transport and from regional sources further away from the sites, by calculating the difference between concentrations measured in Slagslunde and outside of the village.

Main results

A central result of the study is a so-called 'wood smoke source profile', which relates several measures of wood smoke pollution to each other. Wood smoke pollution results in an increased concentration of fine particle mass ($PM_{2.5}$), but also by an increased level of soot, by increased

particle volume, and by an increase in various other compounds. The source profile is a useful tool for interpretation of future measurements.

The contribution of wood smoke to the $PM_{2.5}$ level at the central measuring site in Slagslunde amounts to 2.0 $\mu g/m^3$ as an average for a period of approximately nine weeks during a mild winter.

The report demonstrates how measurements of the particle size distribution are useful for identifying the contribution from various sources, while also describing the influence of wood smoke on the particle size distribution.

During ten days measurements of PAH (Polycyclic Aromatic Hydrocarbons) were carried out. The measurements showed a substantial increase in the level of PAH in the wood stove district compared with the background site. The number of samples is too small to draw conclusions about the level of PAH on a yearly basis.

In various other studies levoglucosan and mannosan are used as indicators for wood smoke. However, the work on the wood smoke source profile presented here demonstrates that measurements of these two substances are not adequate for quantification of the contribution of residential wood combustion to $PM_{2.5}$.

The measurement campaign

Slagslunde is a small village with approximately 400 detached houses, almost all single-storied, surrounded by agricultural areas, and with no local industry. Approximately half of the houses are equipped with wood stoves.

Measurements were conducted at a site centrally in Slagslunde and at a background site approximately 500 m outside the village. Simultaneous measurements with all the equipment running at both sites took place from 23 December 2006 until 26 February 2007. The consumption of wood for combustion in each house was mapped in detail by use of a questionnaire.

The village was situated at a gentle hill. Meteorological measurements were carried out at a mast placed 500 m NW of the village on top of the hill.

During the experimental campaign continuous measurements of a number of air pollution components were conducted: $PM_{2.5}$, particle number and size distribution, CO, NO and NO_x, soot particulates.

In addition to the continuous measurements, during 10 non-consecutive days, 24-hour high volume samples of particles and semi-volatile compounds were collected in order to measure PAH, levoglucosan and mannosan. PAH is carcinogenic, while levoglucosan and mannosan are of interest because they are characteristic for biomass combustion.

In a two-week campaign the relation between indoor and outdoor pollution was examined in two homes: one with, and the other without a woodstove.

Data analysis

An important step in the data analysis was to identify other potential sources than wood combustion.

The purpose of the NO_x (and NO) measurements was to make corrections for the traffic, which is the main source of NO_x, but may also be a disturbing local source of particle number (N) and $PM_{2.5}$. From other studies the N/ NO_x ratio and the $PM_{2.5}$/ NO_x ratio for traffic emissions are approximately known. This characteristic of traffic emissions can be used to assess the traffic contribution to the observed increments. Quality control of the NO_x data after the campaign unfortunately showed that the monitors at both sites had not worked properly except for a period of 23 days. Therefore, it was not possible to make the corrections. Using the NO_x increments measured in the 23 day period it was estimated that a small, but not insignificant part (10-20 %), of the particle number increments can be due to local traffic. For $PM_{2.5}$ it was estimated that the local traffic contributions to the observed increments are almost insignificant (< 5%).

For further investigation of possible disturbing local particle sources the DMPS size spectrum data turned out to be useful. A peak in the increment spectrum around a particle size of 13 nm was sometimes present, but only in periods with certain wind directions. The peak had a positive sign at wind directions around 280°, and a negative sign at wind directions around 140°. These are exactly the directions to the district heating station from the exposed site and the background site, respectively. With other wind directions and when the heating station was turned off in the late night there was no 13 nm peak. Also some episodes with high concentrations of traffic generated particles could be identified by the presence of a peak in the size spectrum around 26 nm.

Based on the analyses of the DMPS size spectrum data it was possible to construct a 'clean data set', for which the increments represent almost only wood combustion; this data set was used for the further analyses. Data measured during the strong traffic episodes and with wind directions coming from the central heating station were excluded from the cleaned data set. The cleaned data set includes 62% of the half-hours compared to the full set. A significant difference between the entire data set and the cleaned data set of the increment is found only for the *particle number* concentration. The reason is that the traffic source and the district heating station both produce many particles, but very little mass, particle volume and soot compared with the wood stoves.

The diurnal variation (hourly averages using the entire cleaned data set) shows elevated concentrations in the late afternoon and evening for the increments of particle volume, soot, $PM_{2.5}$ and particle number. This fits well with the data from the questionnaire study, where the residential wood combustion in Slagslunde was mapped in detail.

The fact that the diurnal variations of $PM_{2.5}$, V and Soot are similar indicates that only one major source (the wood stoves) is the cause of the increments at the site in the village. The diurnal variation of the particle *number* is slightly different with relatively increased levels in the middle of the day, which may be due to traffic emissions.

Source profile for wood smoke

Using the receptor model COPREM a constant 'wood smoke profile' was determined, which gives good predictions for $PM_{2.5}$, V and Soot. The prediction of the particle number N was less satisfying, thus supporting the hypothesis that other local sources than wood smoke contribute to the particle number.

The increments of the PAHs were calculated on the basis of seven 24h samples which were collected simultaneously at the two sites. For these samples all measured values at the background site and at the exposed site were above the detection limit, and data for $PM_{2.5}$, V and Soot were available. A regression analysis was made for the PAH increments as function of the source strengths found by the COPREM model. The correlations between the PAH increments and the increments of $PM_{2.5}$, V and Soot were poor, which is reflected in the high uncertainties on the PAH values in the resulting source profile.

In other studies of pollution from wood combustion the monosaccharide anhydrides levoglucosan and mannosan have been used as indicators for wood smoke, because they are formed precisely by biomass combustion. Increments of levoglucosan and mannosan for nine 24h samples were studied. A woodstove source profile including levoglucosan, mannosan, $PM_{2.5}$, V and Soot was determined. However, levoglucosan and mannosan showed poor correlation with $PM_{2.5}$, V and soot, which is reflected by high uncertainties for levoglucosan and mannosan in the source profile.

A central and useful result of the study is a 'wood smoke source profile' - i.e. a vector which links the three measures $PM_{2.5}$, V and soot to each other, so that from knowledge of the increment of one of these, one can estimate the value of the other two - provided that the source responsible for the increment is wood smoke. However, the increments of the parameters particle number N, PAH and monosaccharide anhydrides do not have a close link to these three measures – they can only be calculated with large uncertainty. Thus, the study indicates that although levoglucosan and mannosan are specific markers for biomass combustion, they are not adequate for quantification of the contribution of $PM_{2.5}$.

The resulting source profile for wood smoke, which indicates average increments due to the wood smoke source in the period corresponding to the cleaned data set were:

$PM_{2.5}$	1,91 ± 0.05	$\mu g m^{-3}$
V	1,78 ± 0.07	$\mu m^3 cm^{-3}$
Soot	2,01 ± 0.08	Mm^{-1}

PAH

The level of PAH at the exposed site was clearly elevated compared to the background site. For benz(a)pyrene (BaP) the concentration was increased by a factor of 2.3. The average BaP concentration at the exposed site was 1.68 ng/m^3. The available data material is much too limited to allow any conclusions about the annual average, but for comparison it

can be noted that the EU target value for BaP is 1 ng/m^3 as an annual average.

Indoor air pollution

Results from the minor campaign where indoor concentrations were measured in two homes are not treated in detail in the present report. However, the following observations can be noted:

- In a house *without* wood stove the level of soot was substantially smaller than the outdoor level.
- In a house *with* woodstove the level of soot was approximately at the same level as outdoor when averaged over a week. However, there is a strong time variation. Whenever a cold stove is lit a very pronounced peak far above outdoor levels occurs. Subsequently, with an active stove, the soot level gradually decreases over 5-7 hours and approximately reaches the outdoor level. When the stove becomes inactive, the level falls below the outdoor level.

1 Introduction

The study described in the present report has been conducted as part of a comprehensive research project entitled *Residential Wood Combustion and the interaction between technology, user and environment* – in short: **WOODUSE**. The project is interdisciplinary and covers many aspects of wood smoke pollution, as it investigates emissions, ambient and indoor pollution levels, health effects and human exposure, social aspects and abatement options. The present report concerns a three-month measuring campaign in the Danish village of Slagslunde, targeted at the study of residential wood combustion.

Many other publications have been produced within the Wooduse project; the project web site http://wooduse.dmu.dk gives an overview of all published material.

The WOODUSE project is supported by the Danish Council for Strategic Research (ENMI-2104-05-0010).

The campaign in question took place in December 2006 - March 2007 in the village of Slagslunde approximately 25 km North of Copenhagen. The campaign comprised measurements of many air pollution components at two sites: an exposed site within the village, and a background site outside of it. The subsequent chapters describe in detail the measurements and the results of data analyses. Two sub-studies were carried out during the campaign. One concerned indoor pollution which is dealt with only very briefly in the present report (section 2.4), whereas more details can be found elsewhere (Olesen et al., 2010b; Toftum, 2010). The other sub-study mapped the emission from woodstoves in the village in considerable detail by use of a questionnaire. Residents of the village were requested to log their use of woodstoves and similar appliances during a six week period (December 16, 2006 - January 28, 2007). This questionnaire-based component of the campaign is described separately (Olesen et al., 2010a).

Previously, studies somewhat similar to the present have been conducted by NERI (Glasius et al., 2006; Glasius et al., 2008; Glasius et al., 2007). However, certain features distinguish the campaign described here.

Compared to previous studies, it was possible in the current campaign to make a more precise determination of the increment from residential wood combustion to concentration in the village, because two monitoring stations were operated simultaneously close to each other. One was located centrally in the village and exposed to many woodstoves, while the other was 500 m outside the village - in contrast to previous studies where background data had to be deduced from a site located several kilometres away from the site

Slagslunde was selected as a suitable site because it is confined in extent, and there are only few sources other than wood combustion. The effect of other sources was expected to be small. This has been confirmed, and

it has been possible to assign a 'flag' to data, describing whether other sources than wood combustion play a role, and subsequently construct a 'cleaned' data set for various analyses. In particular, the present report deduces a "wood smoke profile", which links the three measures $PM_{2.5}$, particle volume V and soot to each other, so that from knowledge of the increment of one of these, one can estimate the value of the other two - provided that the source responsible for the increment is wood smoke. Particle number N does not have a close link to these three measures.

The analysis of results illustrates the usefulness of measurements of size spectra as a sensitive indicator to reveal information about source contributions.

The emission from woodstoves in the village was mapped in detail by use of a questionnaire.

The present report is one among several which present results from the WOODUSE project. Other reports concerning the campaign are the following:

- *Brændefyrings bidrag til luftforurening. Nogle resultater fra projektet WOODUSE* (Olesen et al. 2010b) is a report in Danish (*Contribution to air pollution from residential wood burning. Some results from the WOODUSE project*) which summarises some of the current knowledge on air pollution from residential wood combustion in Denmark, and which also includes model computations based on information from the Slagslunde measuring campaign and the accompanying questionnaire.
- *Brug af brændeovne i Slagslunde. Resultater fra en spørgeskemaundersøgelse inden for projektet WOODUSE* (Olesen et al., 2010a). This report in Danish describes results from the questionnaire-based study, where the use of woodstoves in Slagslunde was mapped with considerable detail for a six-week period.
- A note by Toftum (2010), which presents results from a sub-campaign focussing on the relation between indoor and outdoor pollution (cf. Section 2.4).

The report has the following structure.

Chapter 2 provides a comprehensive description of the measurements.

Chapter 3 presents results and some analyses of the results. A 'cleaned' data set is constructed, comprising data where wood combustion was the only significant local source to air pollution in Slagslunde. Subsequently a 'wood smoke source profile' is derived. This profile is a result of a general nature, and can be used in other contexts.

Chapter 4 contains conclusions.

2 The measurements

2.1 The site

Measurements were made in the small village of Slagslunde, with approximately 400 detached houses, almost all single-storied, surrounded by agricultural areas, and with no local industry. The shape of the populated area is almost a square with a side length of 700 meters. The village is situated on a longish hill with altitude (elevation above see level) ranging from 25 meters at the southern corner to 45 meter at the northern corner. There are no larger towns in the vicinity; except for Copenhagen 20-30 km to the SE.

The village has a small combined power and district heating station using natural gas. This station has a 20 meter high stack and is positioned at the grounds of the public school in the western part of the village. The heat delivered from the heating station is expensive, and many of the inhabitants use wood stoves as substitute or as supplement. Otherwise, heating by oil burner or by electricity is used. A pipeline system delivering natural gas to the individual houses has not been implemented, because this would counteract the idea of the district heating system. Approximately half of the houses are equipped with wood stoves.

All air pollution monitoring instruments were placed in two mobile monitoring stations. One station was exposed to smoke from woodstoves at a site in the middle of the southern part of the village, and the other station was placed for background reference 500 meters outside the village to the WNW at 45 meter altitude (see Figure 2.1). The background station was placed on a grass field close to a farm with only one occupying resident and without heating of any kind except for the natural heat produced by the animals in the cattle shed.

At both stations the sampling height was about 3 m above ground level. Simultaneous measurements with all the equipment running at both stations took place from 23 December 2006 until 26 February 2007, but some measurements continued until 19 March.

The traffic near the sampling sites is modest. Most important as local particle sources are the connecting secondary roads closest to the monitoring stations. In average the traffic density on these roads is 1260 vehicles per day at the exposed site, and 1890 vehicles per day at the background site (ref. Maria Matzen, Miljøcenteret, Egedal Kommune).

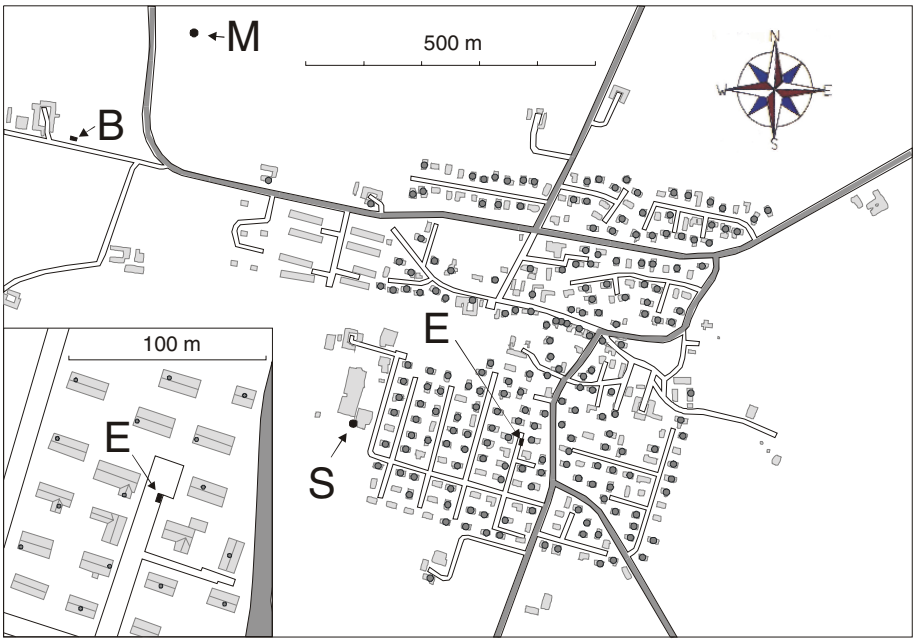


Figure 2.1. Map of the village Slagslunde showing connecting secondary roads (dark), local roads (white), and buildings. The dark circles indicate houses with wood stoves. The letters indicate the positions of the exposed site (E), the background site (B), the meteorological mast (M), and the 20 meter high stack of a district heating station (S). The position of the wood stove chimneys close to the exposed site is shown in the inserted magnification.

2.2 Meteorological measurements

Meteorological measurements were performed at a mast placed 500 m NW of the village on the top of the gentle hill with the village. The mast had a height of 7 m with its base 50 m above sea level. It was equipped with a sonic anemometer (Metek model, USA-1) placed on top of the mast, temperature sensors at two heights, and a pyranometer for global radiation measurements (Soldata model SPC80). Sonic data were sampled with 10 Hz. These data were used to provide a basis for dispersion calculations (described by Olesen et al., 2010b).

2.3 Measurements of air pollution components

During the experimental campaign, continuous measurements of a number of air pollution components were conducted: $PM_{2.5}$, particle number and size distribution, CO, NO and NO_x, soot particulates.

In addition to the continuous measurements, during 10 non-consecutive days, 24-hour high volume samples were collected in order to detect PAH, levoglucosan and mannosan.

Furthermore, a sub-campaign was conducted during two weeks in order to examine the relation between indoor and outdoor pollution in two homes: one with, and the other without a woodstove. This sub-campaign is briefly discussed in section 2.4, while more details are found in other reports (Olesen et al., 2010b; Toftum, 2010).

2.3.1 Measurements of $PM_{2.5}$

Particle measurements of $PM_{2.5}$ were carried out using TEOM instruments (Rupprecht and Patashnick, NY, USA) operated at 40 °C to dry the aerosol. Besides removing the water the elevated temperature results in loss of volatile compounds (especially ammonium nitrate), and in general $PM_{2.5}$ values obtained with TEOM are lower than results obtained with gravimetric or beta-attenuation methods (Hitzenberger et al., 2004; Hauck et al., 2004). In other contexts TEOM measurements are sometimes 'corrected' in order to match gravimetric measurements. Such a correction is not applied to the data presented here, because ammonium nitrate is not directly emitted by wood stoves. However, some unknown losses of other volatile compounds from the woodstoves may still influence the $PM_{2.5}$ results. The data were averaged for each half-hour. Due to delayed evaporation of volatile material, negative values for half-hourly values of $PM_{2.5}$ can occur. Such values are retained in the data set to avoid bias on calculated average values.

2.3.2 Measurements of particle number and particle size distribution

The particle size distributions (10-700 nm) were measured with a Differential Mobility Particle Sizer (DMPS) using a Vienna type Differential Mobility Analyser (Winklmayr et al., 1991) coupled to a Condensation Particle Counter (TSI 3010) and employing a re-circulating flow system (Jokinen and Makela, 1997). The scanning time for each size spectrum was 2 min, using alternating up- and down-scans. By integrating over the particle size distribution the total particle number and particle volume in the range 10-700 nm was calculated assuming spherical particles with geometrical diameters equal to the mobility diameter for the volume calculation. The data were averaged for each half-hour.

2.3.3 Measurements of CO, NO and NO_x

Half-hourly averages of NO and NO_x were measured with API M 200A (San Diego, Califonia) monitors using chemiluminescence detection. Half-hourly averages of CO were measured using API M300 monitors using infrared absorption.

2.3.4 Measurements of soot particles

The light absorption by particles collected on a filter was measured by custom built Particulate Soot Absorption Photometers (PSAP) following the design described in Bond at al. (1999). The PSAP measures the absorption of light using green light, 550 nm, passing through a glass fibre filter (Pallflex E70-2075W) that is continuously loaded with particles collected from ambient air. The instrument also provides a reference signal allowing corrections for electronic noise and drift. The measurement and the reference signals are recorded continuously and the flow is kept constant by a critical orifice. The filter is rotated automatically or has to be replaced manually if the soot loading has reduced the signal to about 30%-50% of the reference signal. Several laboratory tests and calibrations have been performed (Andersen, 2006) showing a reliable performance of the instruments. The light absorption coefficient (optical attenuation per length in ambient air) is measured in 15-minute intervals and averages are calculated for half-hour intervals. If needed, the light absorption coefficient can be converted to a soot mass concentration dividing by an

absorption cross section per unit mass of 7.5 ± 1.2 m^2g^{-1} recommended in the review of Bond et al. (2006), or by the value of 12.7 m^2g^{-1} originating from an intercalibration with a Ambient Carbon Particle Monitor (ACPM R&P inc.) at a street location in Copenhagen (Andersen, 2006). It should be noted, however, that such conversion refers to the mass of *soot exclusively*. $PM_{2.5}$ comprises other compounds than soot; therefore the listed factors cannot be used to quantify $PM_{2.5}$. In section 3.7 it is quantified how an increment in soot concentration caused by woodstoves is accompanied by a corresponding increase in $PM_{2.5}$ concentrations.

2.3.5 Measurements of PAH

During 10 non-consecutive days, 24-hour samples were collected on filter in order to detect PAH, levoglucosan and mannosan.

Particulate and semivolatile compounds were sampled on a quartz microfiber filter type QM-A, 102 mm diameter (Whatman) followed by two cylindrical polyurethane foam (PUF) plugs (6 cm diameter and 5 cm long with a density of 0.02 g cm^{-3}) positioned in series. The sampling flow rate was 130 l min^{-1}. Samples were collected over a 24 hours period from about 9 a.m. Samples were stored at –18 °C until analysis. Only the filters were analyzed. Half of the quartz filter was analysed for PAH, while a quarter of the filter was analysed for levoglucosan and mannosan.

For the analysis of PAH the samples were spiked with deuterium-labelled PAH standard prior to extraction in order to determine analytical recovery. Filters were extracted by sonication with dichloromethane. The solvent was exchanged to hexane and the extracts were cleaned on silicic acid glass columns (1000 g/6 ml). The columns were first eluted with 5 ml hexane followed by 5 ml dichloromethane. The dichloromethane fraction was collected for the analysis of PAHs. The solvent was further reduced to dryness and re-dissolved in toluene containing deuterium-labelled PAH as internal standards. Samples were analyzed on a TurboMass GC-MS (Perkin Elmer, Norwalk, CT, USA) operated in selected ionization mode (SIM) under the following conditions: capillary 30 m x 0.25 mm i.d. capillary column DB-5, 0.25 µm film thickness. Samples were injected in splitless mode. The temperature program was as follows: 90 ºC for 2 minutes, 20 ºC/min to 240 ºC, hold for 5 minutes, 3.5 ºC/min to 280º C, 25 ºC/min to 300 ºC, hold for 12 minutes. Concentrations of individual PAH in samples were corrected for recovery by a deuterium-labelled PAH standard with the closest molecular weight.

2.3.6 Measurements of levoglucosan and mannosan

For the analysis of the monosaccharide anhydrides (levoglucosan and mannosan) a quarter filter was spiked with the recovery standard $^{13}C_6$-levoglucosan. The filter was extracted 3 times with 50 ml methanol in an ultrasonic bath for 30 minutes each time. The combined extracts were evaporated to a volume of 1 ml and filtered through a nylon filter. Analysis was performed with liquid chromatography-mass spectrometry (LC-MS) equipped with electrospray ionization (ESI) operated in positive mode.

2.4 Indoor versus outdoor concentrations

Within the Slagslunde measuring campaign a minor sub-campaign was conducted in order to study the effect of a woodstove on indoor pollution.

The present section briefly outlines the measurements conducted, while more information on results can be found in a report in Danish by Olesen et al. (2010b), which includes a presentation of indoor-outdoor concentration results. Further, there is a report with focus on air flow measurements (Toftum, 2010).

2.4.1 Method

Indoor and outdoor pollution was measured simultaneously for one week in two houses: one with a wood stove (March 6-12), and another without a stove (March 12-18). The wood stove was a Lotus stove from 1997. The measuring site in the village was located close to both houses (at a distance of 20 m to the house with wood stove, and 50 m to the house without). Focus was on pollution with soot, because soot is more specific for wood smoke than $PM_{2.5.}$ Soot measurements were performed with PSAP with a time resolution of 15 minutes as described in section 2.3.4. The residents of the houses logged activities such as cooking, cleaning, opening windows etc.

Further, measurements were conducted of outdoor to indoor transport of air as well as between living room and bedroom and between bedroom and living room in each residence using a passive tracer gas technique. These measurements provided weekly averages. The air flow measurements and results are described in more detail by Toftum et al. (2010).

Results for indoor-outdoor sub-campaign

Results of the indoor-outdoor sub-campaign are not presented here in any detail. However, a few main results can be noted:

- In the house *without* wood stove the level of soot pollution is substantially smaller than the outdoor level.
- In the house *with* woodstove the level of soot concentration is approximately at the same level as outdoor when averaged over a week. However, there is a strong time variation. Whenever a cold stove is lit a very pronounced peak far above outdoor levels occurs. Subsequently, with an active stove, the soot level gradually decreases over 5-7 hours and approximately reaches the outdoor level. When the stove becomes inactive, the level falls below the outdoor level.

3 Results and 'data cleaning'

The winter of 2006/2007 in Denmark was mild most of the time with wind from south or west. Only in few periods the daily average temperature dropped below the freezing point. In the colder periods the typical wind direction was north or east. These shifts between periods with mild oceanic air and periods with colder continental air are typical for the winter weather in Denmark. The temperature and wind data (half-hour averages) from the meteorological station are shown in Figure 3.1, Figure 3.2, and Figure 3.3.

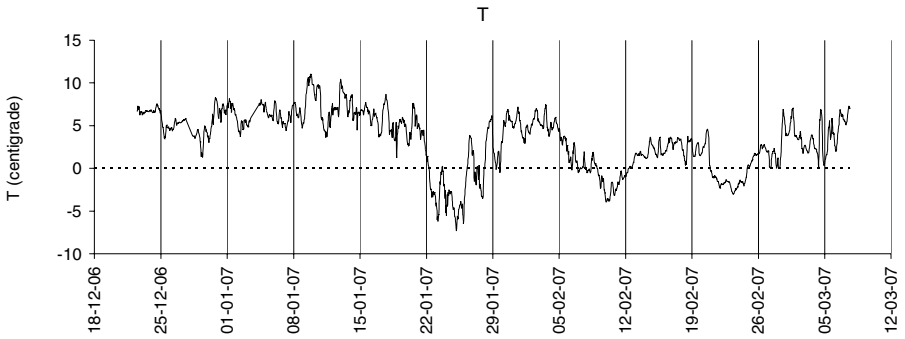


Figure 3.1. The temperature (T) as half-hourly averages measured during the campaign at Slagslunde.

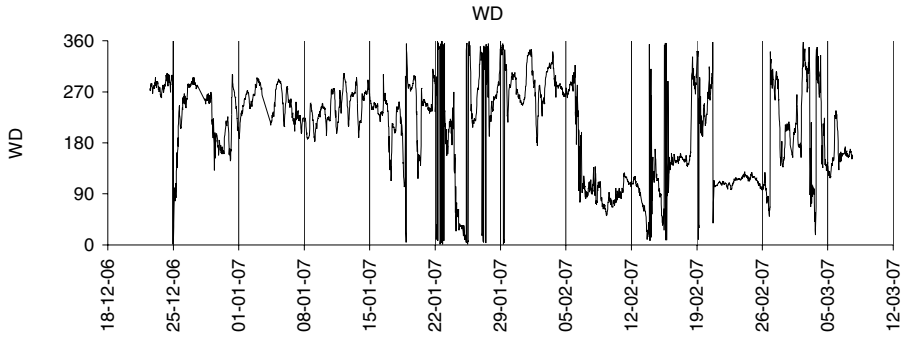


Figure 3.2. The wind direction (WD) in degrees during the campaign (N = 0, E = 90).

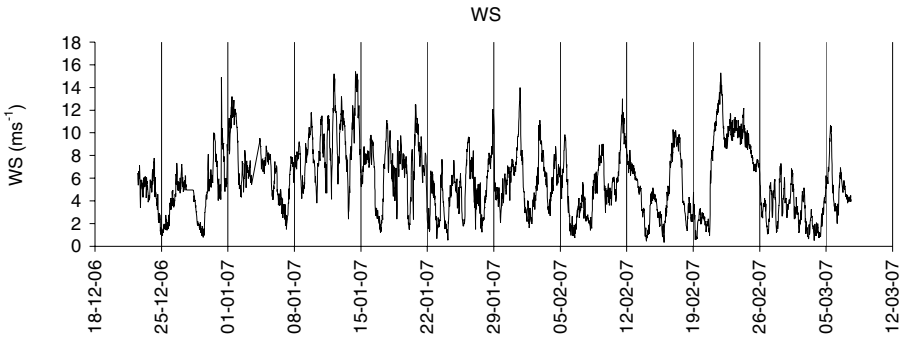


Figure 3.3. The wind speed (WS) during the campaign.

As the objective of the study was to estimate the influence of local wood combustion on the air quality it was important to identify other possible sources and to quantify or eliminate their contributions. By means of the setup with two closely located measuring stations the regional and long-range influence can be eliminated by calculating the increments, i.e. the concentration of the pollutants at the exposed site minus the concentration at the background site. The influence of local traffic was expected to be small, because there are no major roads with dense traffic in the vicinity of the village. There are only some connecting secondary roads in a distance of 50-100 meter from each site with a number of 1000-2000 vehicles per day.

In the following, we present an exploratory analysis of results. The main intention is to identify contributions from various sources, so we can isolate and quantify contributions from domestic wood burning. The approach illustrates how a combination of simultaneous measurements of several pollution components, as well as particle size measurements, is useful in identifying sources.

3.1 NO_x results and traffic contribution

The purpose of the NO_x measurements was to verify that the traffic is not an important local source. Quality control of the NO_x data after the campaign unfortunately showed that the monitors at both sites had not worked properly in long periods during the campaign. Simultaneous, quality approved measurements were only obtained for 23 days in the periods from 21 December to 27 December, and from 3 January to 20 January.

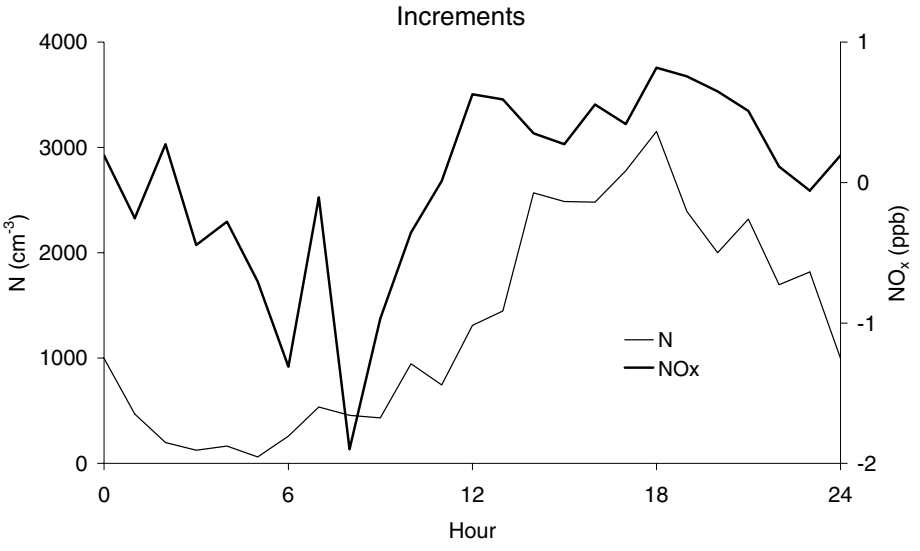


Figure 3.4. The average diurnal variation of the increments (data from the exposed site minus simultaneous data from the background site) of particle number (N) and NO_x during part of the campaign.

The average diurnal variation of the increments (data from the exposed site minus simultaneous data from the background site) of particle number (N) and NO_x during these 23 days is shown in Figure 3.4. From other studies (Wåhlin, 2008) we know that for traffic emissions the N/NO_x ratio and the $PM_{2.5}/NO_x$ ratio are approximately 300 cm^{-3}/ppb and 0.08 μgm^{-3}/ppb, respectively. This characteristic of traffic emissions can be used to assess the traffic contribution to the observed increments. By multiplying the NO_x increments, which are mostly in the range between -1 ppb and +1 ppb, with the N/NO_x ratio we get an estimate of ±300 cm^{-3} for the local traffic contributions to the particle number increments. As the measured diurnal variation of the number concentration is in the range 0 – 3000 cm^{-3} we conclude that a small, but not insignificant part (10-20 %) of this variation can be due to local traffic. For $PM_{2.5}$ we get an estimate of only ±0.08 μgm^{-3} for the local traffic contributions to the increments of $PM_{2.5}$. This is a small number compared to the observed increment of 2 $\mu g/m^3$ for $PM_{2.5}$ (see Table 3.1 later).

3.2 Analysis using particle size distributions

For further investigation of possible disturbing local particle sources the DMPS size spectrum data turned out to be useful. The detailed particle size distributions contain a lot of data, but a more simple description can be achieved by fitting the size spectra with a small number of log-normal modes. A number of four modes were chosen, and the median diameter and width of each mode were adjusted to give the best overall fit by a least squares method to all the collected size spectra. The shape of the resulting modes with median diameters at 13 nm, 28 nm, 65 nm, and 167 nm, respectively, are shown in Figure 3.5 and Figure 3.6 together with the fitted averages of the size distributions over the entire campaign.

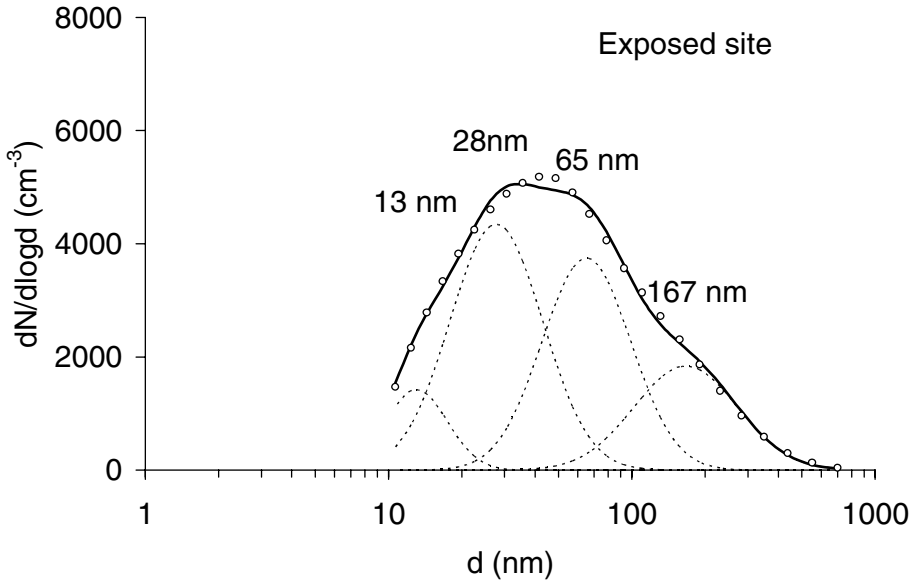


Figure 3.5. The average of the size distributions at the exposed site in the village over the entire campaign (small circles) and the contributions from four fitted modes (the four dotted curves; their sum is a full-drawn curve). The contributions can be read as the areas below the curves, where the unit on the d-axis is one decade (three decades are shown).

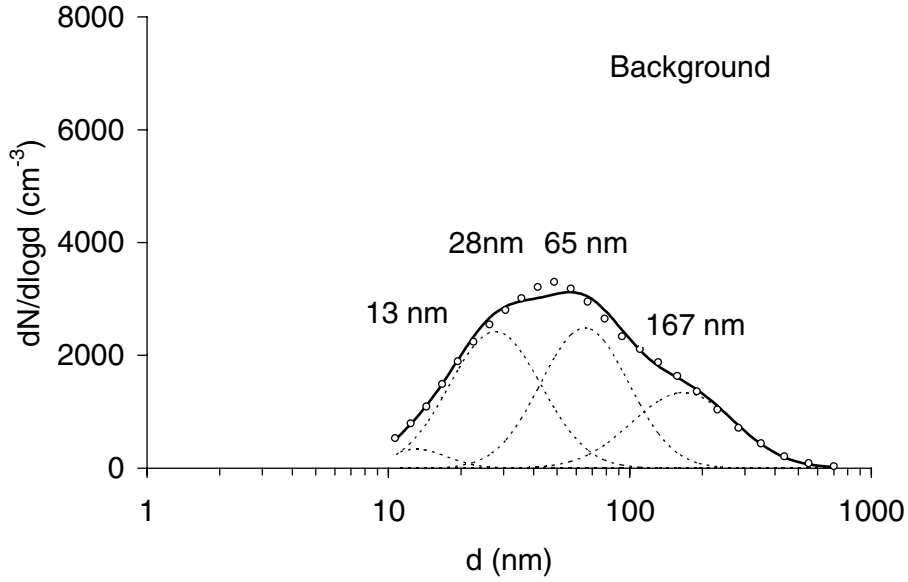


Figure 3.6. The average of the size distributions at the background site outside the village over the entire campaign and the contributions from the same modes as in Figure 3.5.

The time series of the increments of the contributions from each mode to the particle number are shown in Figure 3.7, Figure 3.8, Figure 3.9, Figure 3.10, and Figure 3.11. The increments of $PM_{2.5}$ are shown in Figure 3.12. The circles indicate periods or occasions where the influence from other sources than wood combustion seems to be important. In the period 21 February – 25 February the 13 nm mode and especially the 28 nm mode were dominating. During this period there was a fresh to strong breeze from east with temperatures below the freezing point, as can be seen in Figure 3.1, Figure 3.2, and Figure 3.3. The temperature and wind speed may be favourable for nucleation of volatiles in the exhaust from the traffic passing the nearest point of the local road 70 meter to the east of the exposed (village) site (see Figure 2.1). The diurnal variation of the 28 nm mode increment for this period is shown with more details in Figure 3.9. The high values during daytime and the low values after the afternoon rush hour are indeed typical for traffic. Some similar episodes with an opposite sign are seen during some few hours of the 5 January and 12 January (Figure 3.8). Here the particle source has not been identified, but a probable explanation is that some tractor or truck was running idle close to the background station during these hours.

During the afternoon on 25 January the wind was coming from north and was accompanied for a few hours (14:30-18:00) with high values of the 65 nm mode particles at the site in the village (Figure 3.10). The source of these particles has not been disclosed, but open burning of garden refuse is a probable explanation.

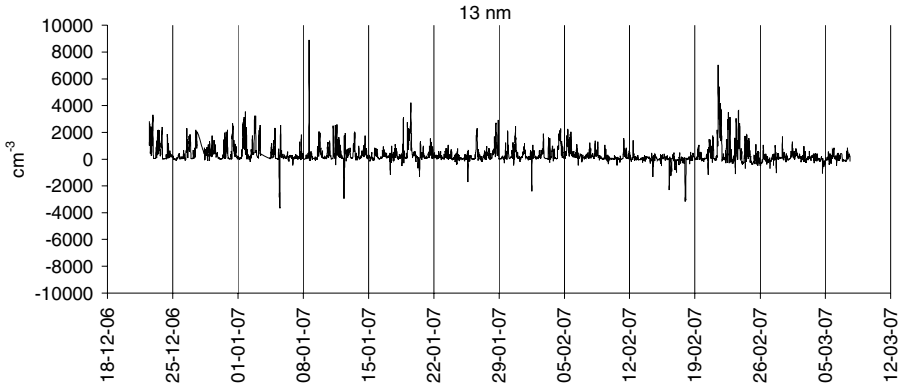


Figure 3.7. The contributions from the 13 nm mode to the total particle number increment during the entire campaign.

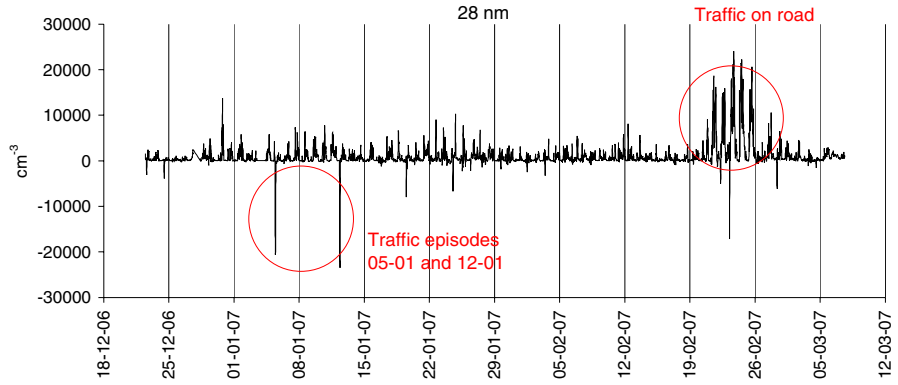


Figure 3.8. The contributions from the 28 nm mode to the total particle number increment during the entire campaign.

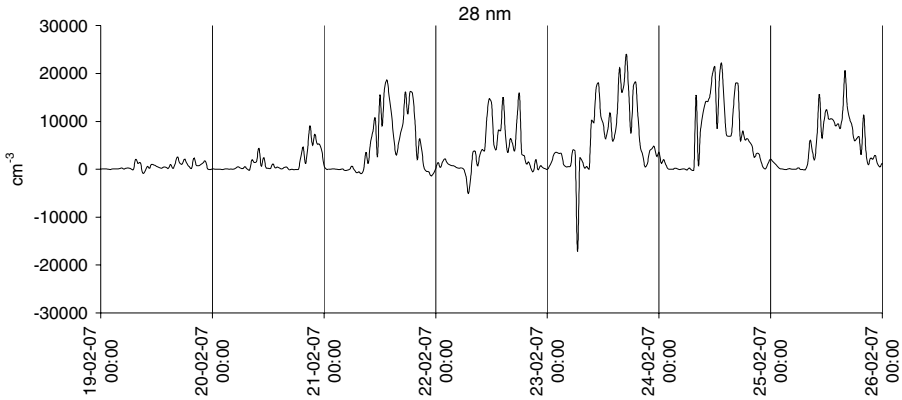


Figure 3.9. The contributions from the 28 nm mode to the total particle number increment during the week 19.02.07-25.02.07.

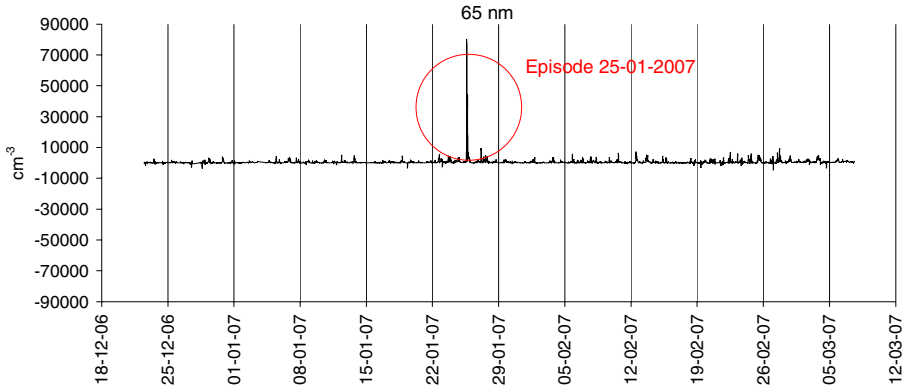


Figure 3.10. The contributions from the 65 nm mode to the total particle number increment during the entire campaign.

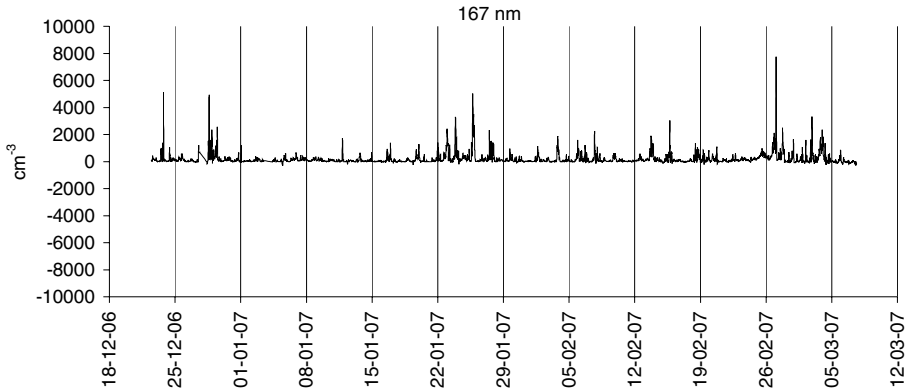


Figure 3.11. The contributions from the 167 nm mode to the total particle number increment during the entire campaign.

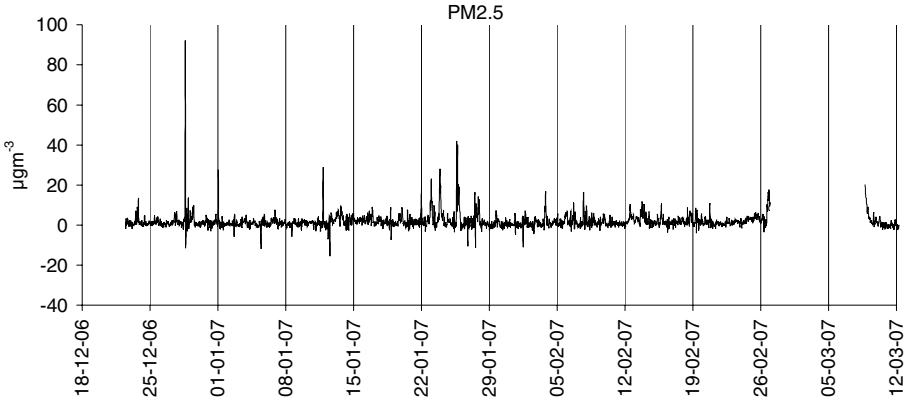


Figure 3.12. The $PM_{2.5}$ increment during the entire campaign on a half-hourly basis

The dependence of the increments of the particle modes on the wind direction is shown in Figure 3.13, Figure 3.14, Figure 3.15, and Figure 3.16. The 13 nm and 26 nm traffic particles coming from the east at the exposed site in the period 21 February – 25 February are seen as a peak in the range 90°-130°. Also the two short traffic episodes at the background site are seen as a single negative peak because the wind was from the same direction (290°) at both occasions. Further, the episode with 65 nm particles coming from the North (350°) can be seen as a peak.

In addition to these peaks, which can be explained by traffic and probably burning of garden refuse for the 65 nm particles, two prominent peaks are seen in the wind direction distribution of the 13 nm particles. One peak with positive sign at wind directions around 280°, and one with negative sign at wind directions around 140°. These are exactly the directions to the district heating station from the exposed site and the background site, respectively. A separate band of points along the axis exists in Figure 3.13 below the peak at 280° (the separation is not easy to see in the printed version of the report). The explanation for the existence of these points is that the gas turbines at the district heating station were turned off during the night-time hours.

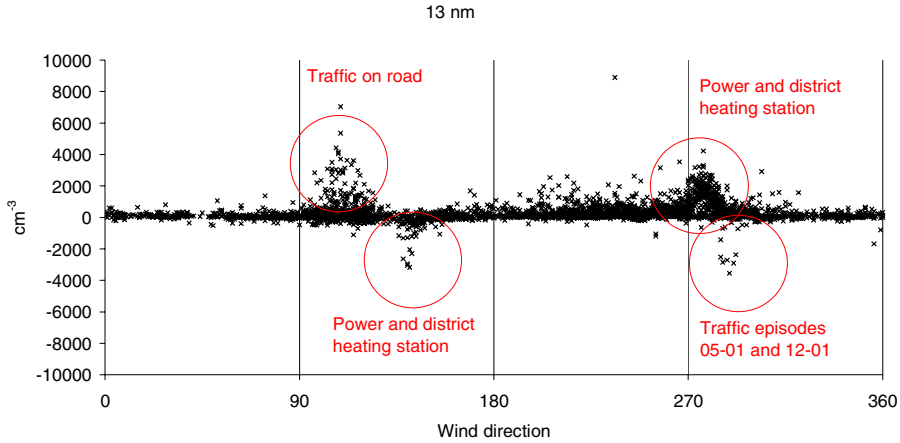


Figure 3.13. The contributions from the 13 nm mode to the total particle number increment as a function of the wind direction.

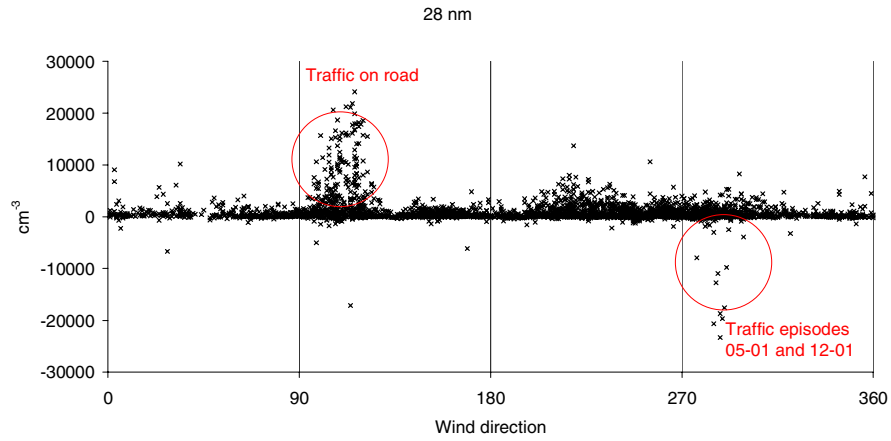


Figure 3.14. The contributions from the 28 nm mode to the total particle number increment as a function of the wind direction.

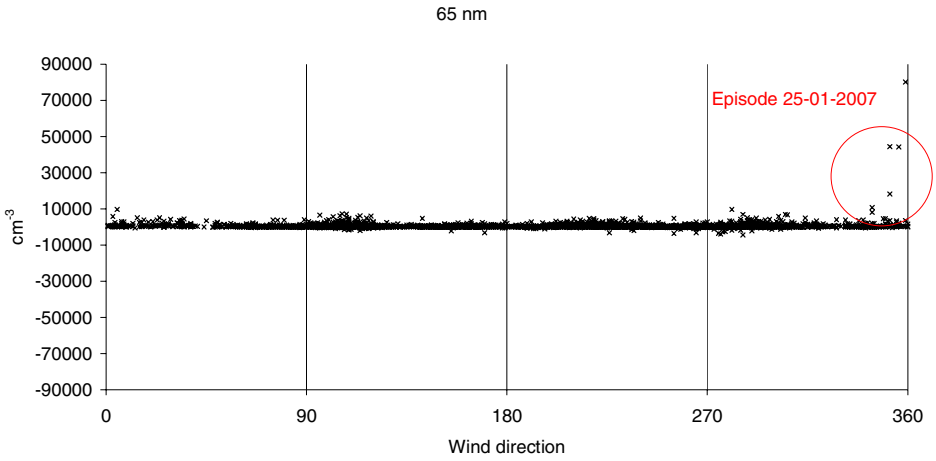


Figure 3.15. The contributions from the 65 nm mode to the total particle number increment as a function of the wind direction.

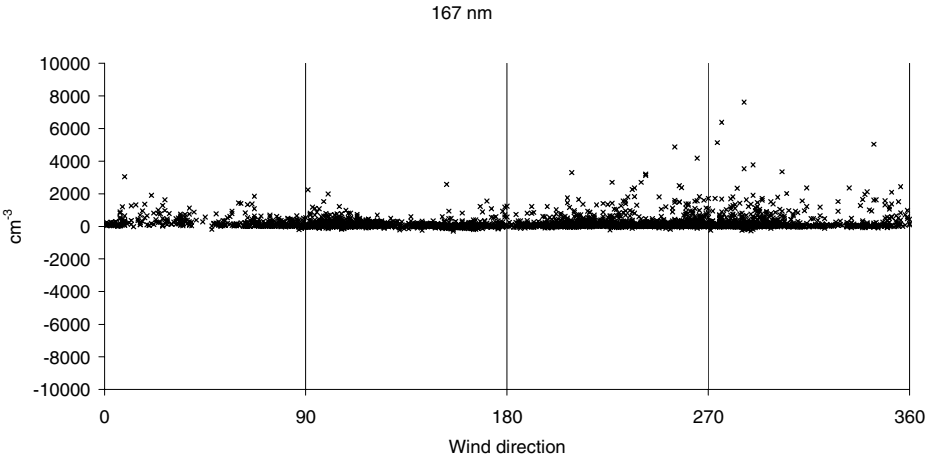


Figure 3.16. The contributions from the 167 nm mode to the total particle number increment as a function of the wind direction.

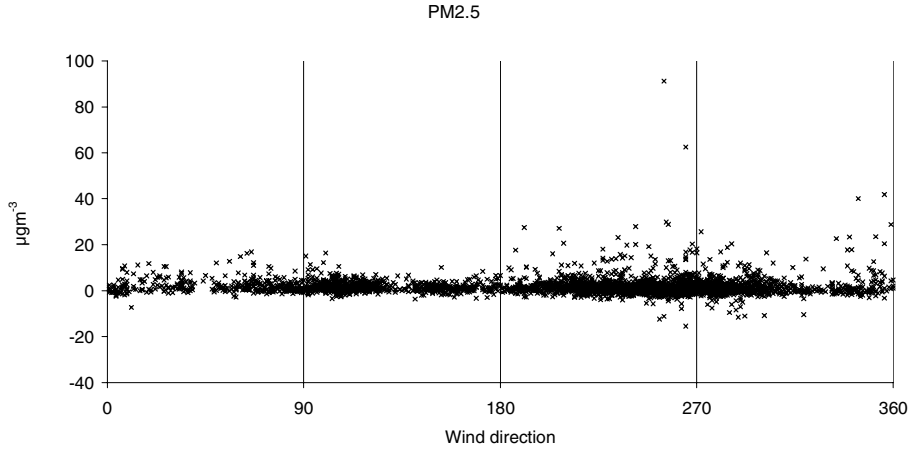


Figure 3.17. The $PM_{2.5}$ increment as a function of the wind direction.

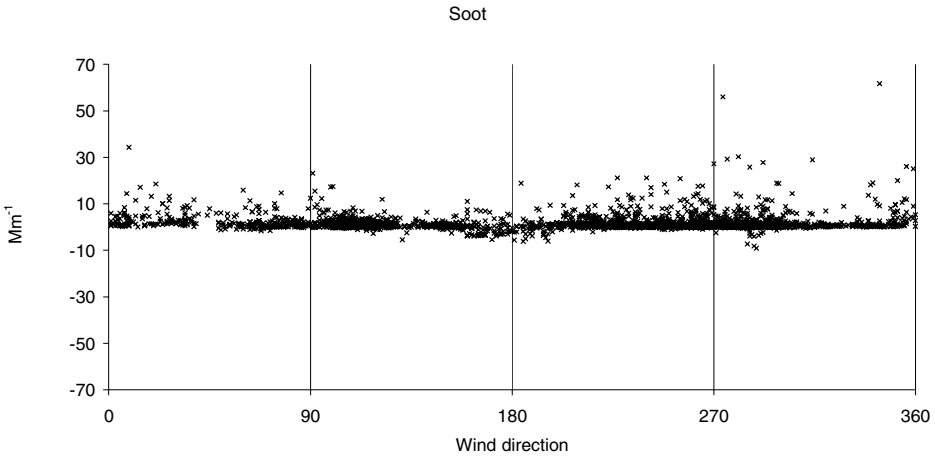


Figure 3.18. The soot increment as a function of the wind direction

3.3 Cleaned data set

The wind direction dependence of the $PM_{2.5}$ and the soot increments are shown in Figure 3.17 and Figure 3.18. None of the peaks in Figure 3.13 - Figure 3.16, which were ascribed to sources like local traffic and the district heating station, are seen in these figures. This fact illustrates the usefulness of measurements of size spectra as a sensitive indicator to reveal influence about source contributions. It is clear, however, that traffic and the district heating station do not contribute much to the $PM_{2.5}$. Based on the analyses above it is possible to construct a 'clean data set', which represents wood combustion; this data set is used for further analyses in the next sections. Data are from the period with simultaneous measurements, i. e. 23 December - 26 February. The following episodes have been excluded from the 'cleaned data set': the short time episodes on 5 January, 12 January and 25 January; all data when the wind direction was from the district heating station at either of the two sites (265°-290° and 130°-150°); all data when the wind was in the direction (90°-130°) where a clear influence from traffic was seen in a period in February. Table 3.1 and Figure 3.19 - Figure 3.26 show results for the full data set and the cleaned data set. Recall that the cleaned set represents only the local emissions in the wind sectors in the ranges 290°-90° and 150°-265° around the exposed site in the village.

Table 3.1 summarises information on averages. The cleaned data set includes 62% of the half-hours compared to the full set. Note that the data underlying the $PM_{2.5}$ values include occasional negative half-hourly values of $PM_{2.5}$, and that $PM_{2.5}$ data are given as uncorrected values measured by TEOM; accordingly, they are not directly comparable with the limit values.

A significant difference between the entire data set and the cleaned data set of the increments (Table 3.1) is found only for the *particle number* concentration. The reason is that the traffic source and the district heating station both produce many particles, but very little mass and soot compared with the wood stoves.

Table 3.1. Averages of $PM_{2.5}$, Soot (light absorption), particle number in the range 10-700 nm (N) and particle volume in the range <0.7 µm (V), at the background site (B), at the exposed site in the village (E), and for the increment E minus B (E-B).

	$PM_{2.5}$ (B)	$PM_{2.5}$ (E)	$PM_{2.5}$ (E-B)	Soot (B)	Soot (E)	Soot (E-B)	N (B)	N (E)	N (E-B)	V (B)	V (E)	V (E-B)	No. of halfhours
	$\mu g m^{-3}$			Mm^{-1}			cm^{-3}			$\mu m^3 cm^{-3}$			
Full data set	8.5	10.4	2.0	3.5	5.3	1.8	2931	4838	1908	5.4	7.0	1.6	2885
Cleaned data set	7.3	9.4	2.0	2.9	4.8	1.9	2537	4037	1500	4.5	6.2	1.6	1792

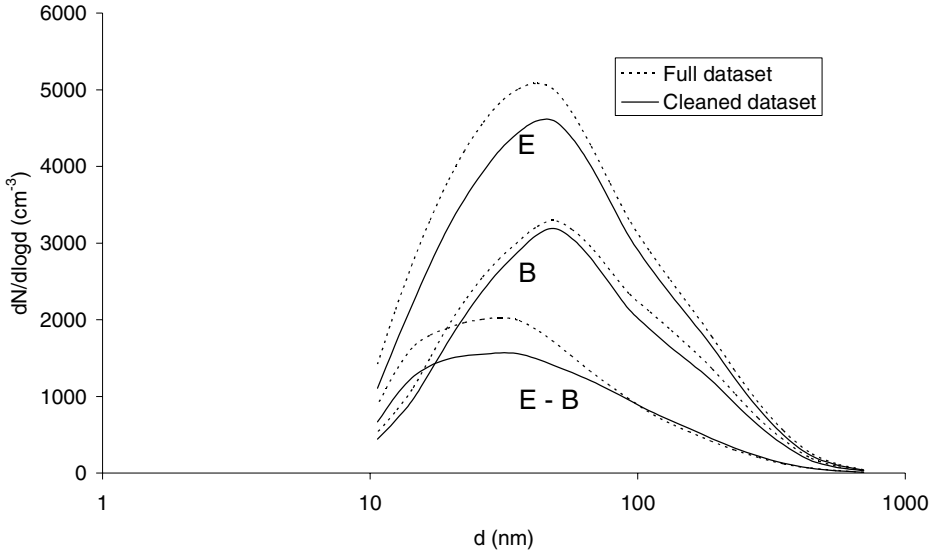


Figure 3.19. The average size distributions at both sites (E and B) and the increment (E-B), both using the full data set and the cleaned data set.

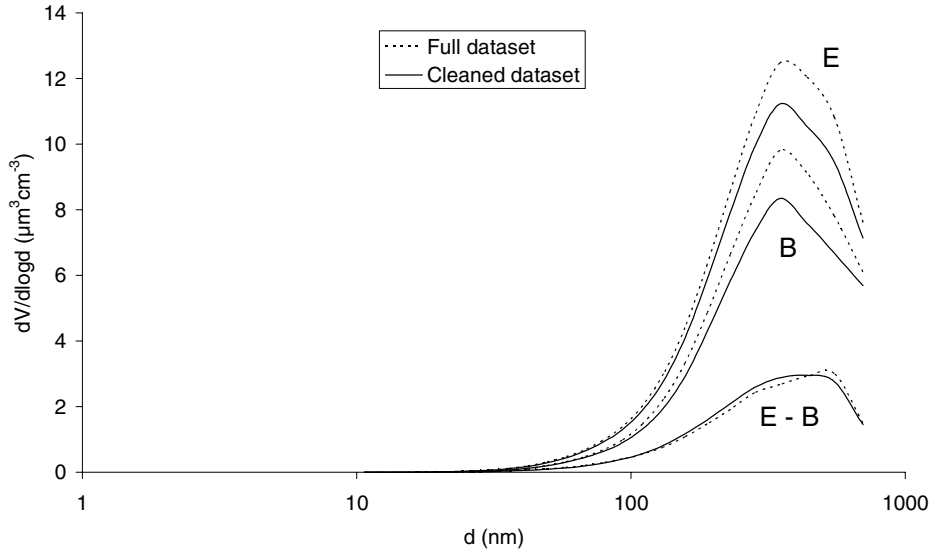


Figure 3.20. The average size distributions converted to volume by multiplication with $\pi/6 \quad d^3$ at both sites (E and B) and the increment (E-B) both using the full data set and the cleaned data set

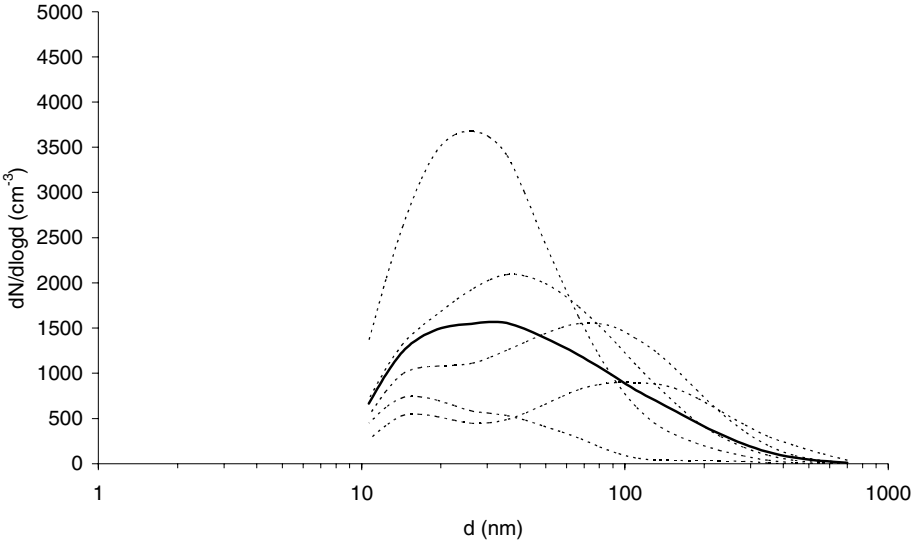


Figure 3.21. The full-drawn line shows the average increment of the particle size distribution (cleaned data set). The five broken lines each represent one fifth of the data.

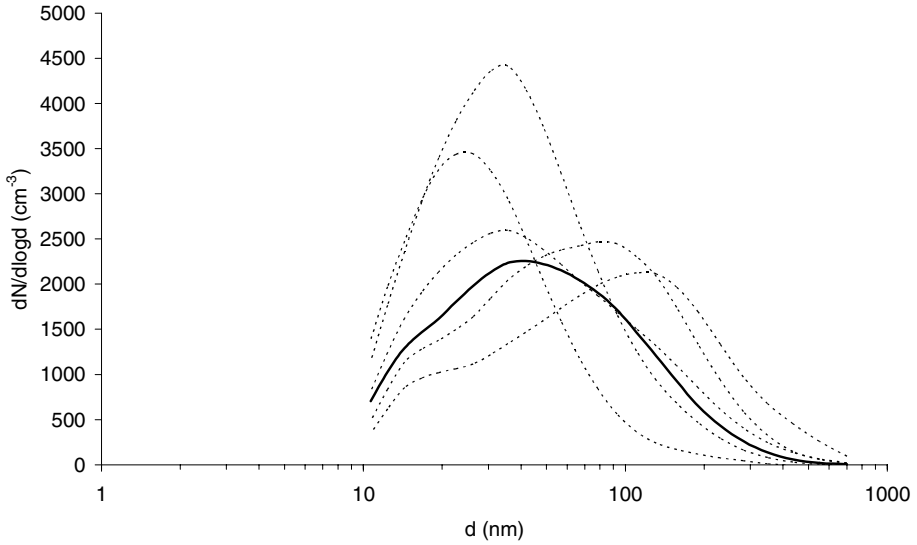


Figure 3.22. Same principle as Figure 3.21 but only with data from the evening period (18-24).

The average particle size distribution of the increment shown in Figure 3.19 does not reveal whether the shape of the distribution varies with time. To give a better impression, the spectra were classified in the following way:

For each half-hour of the cleaned data set the V/N-ratio (the integrated particle volume divided by the integrated particle number) was determined.

Next, the half-hours were sorted according to the V/N-ratio and grouped in five classes, each with the same number of half-hours. Thus, half-hours have been classified according to the dominating particle size. The average size distribution for each of the five classes is plotted together with the total average in Figure 3.21.

The same principle is used for the plot in Figure 3.22, but only with data from the evening (hours 18-24). It is expected that in Figure 3.22 (evening data) the influence from traffic is less than in Figure 3.21, and that the

signal is mainly representative of wood smoke. It is seen that in the evening, the two modes representing the largest particles play a larger role than they do for the all-day data. Further discussion is found in section 3.7.

3.4 Diurnal variation

The diurnal variation (hourly averages using the entire cleaned data set) in the background and for the increments of particle volume, soot, $PM_{2.5}$ and particle number are shown in Figure 3.23 - Figure 3.26.

As an interesting reference, Figure 3.27 is included. This figure is based on data from the questionnaire study, where the residential wood combustion in Slagslunde was mapped in detail. The graph shows the amount of wood burnt in the entire village of Slagslunde. The graph represents an estimate based on all available information from the questionnaire (for details see Olesen et al., 2010a). Note that a correct curve would probably be smoother than the one shown, but the time resolution of the information is not as high as for concentration measurements - the questionnaire operates with five periods each day, not with hourly data.

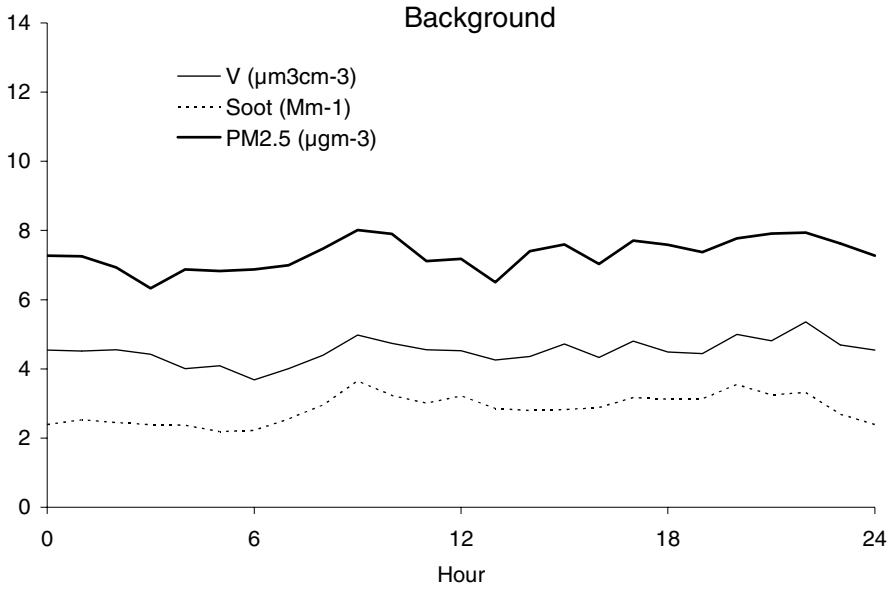


Figure 3.23. The diurnal variation at the background site of the particle volume concentration (V), soot (light absorption) and $PM_{2.5}$. Data from the cleaned data set.

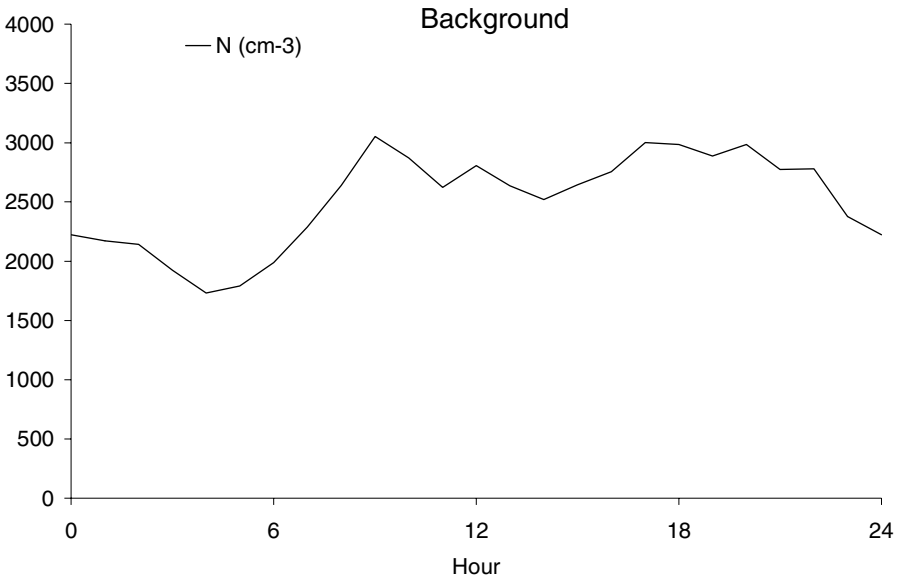


Figure 3.24. The diurnal variation at the background site (B) of the particle number concentration. Cleaned data set.

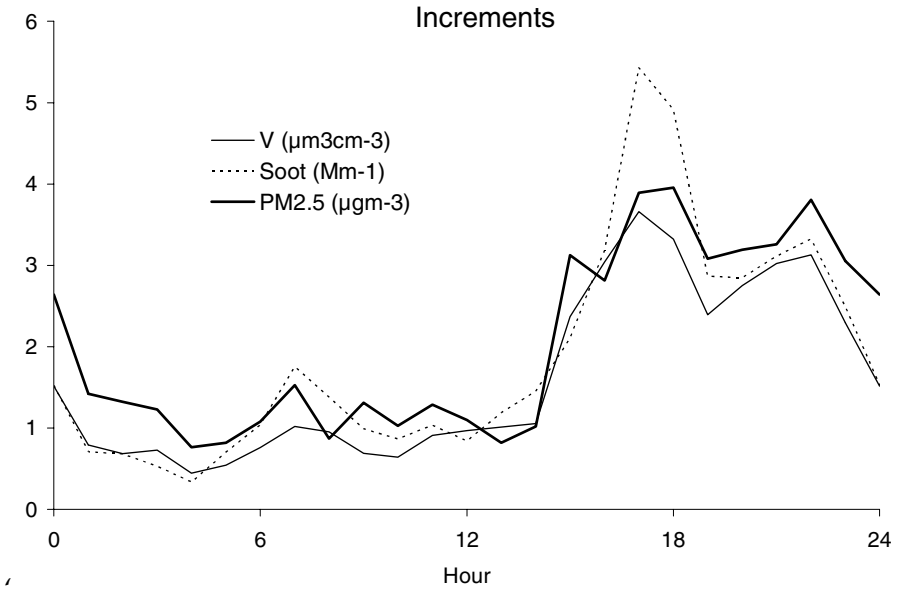


Figure 3.25. The diurnal variation at the exposed site (E) of the increments (E minus Background) of the particle volume concentration (V), soot (light absorption) and $PM_{2.5}$. Cleaned data set.

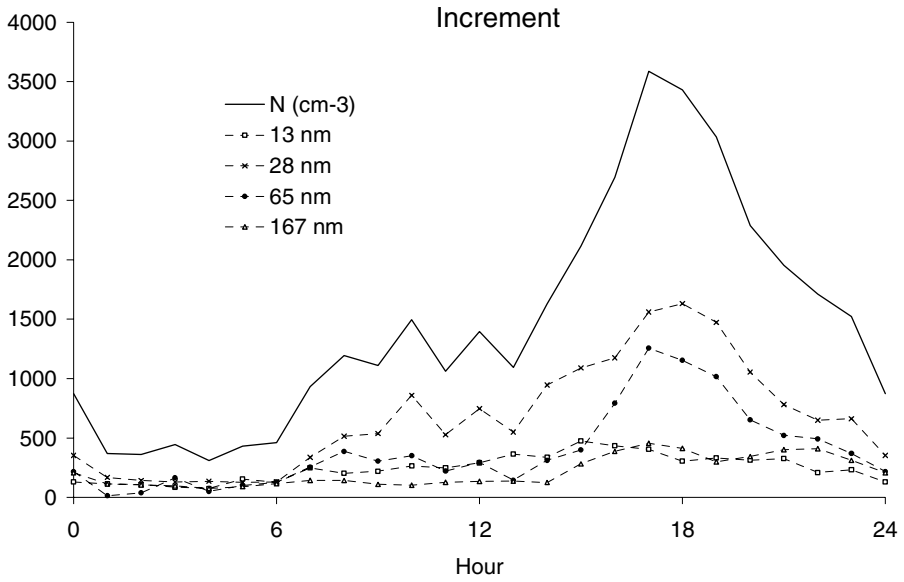


Figure 3.26. The diurnal variation of the increment of the particle number concentration (N). Cleaned data set.

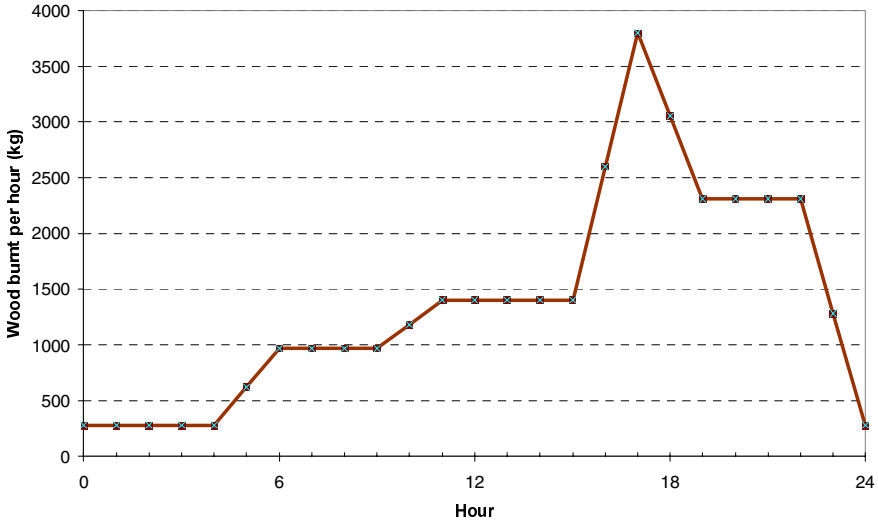


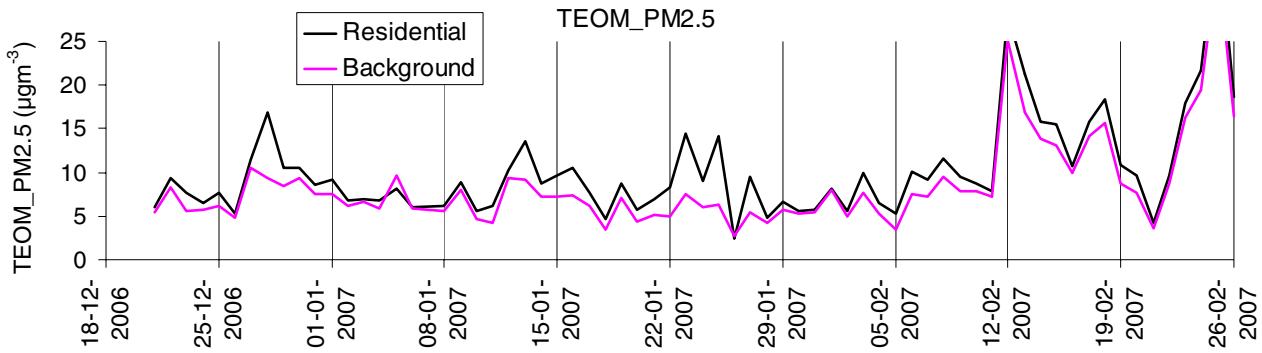
Figure 3.27. Wood burnt in the entire village of Slagslunde by hour of day. An average for the period of the questionnaire (December 16 - January 28). The values are estimated from the responses to the questionnaire (details in Olesen et al., 2010a). The value at 17 hours represents combustion between 5 and 6 pm...

3.5 Results for $PM_{2.5}$

Some results for the $PM_{2.5}$ increment on a half-hourly basis are shown in the previous section (Figure 3.12, Figure 3.17, Figure 3.25).

Figure 3.28a shows a time series of $PM_{2.5}$ concentrations at the exposed site and at the background site, based on daily average values. Figure 3.28b shows the increment.

(a)



(b)

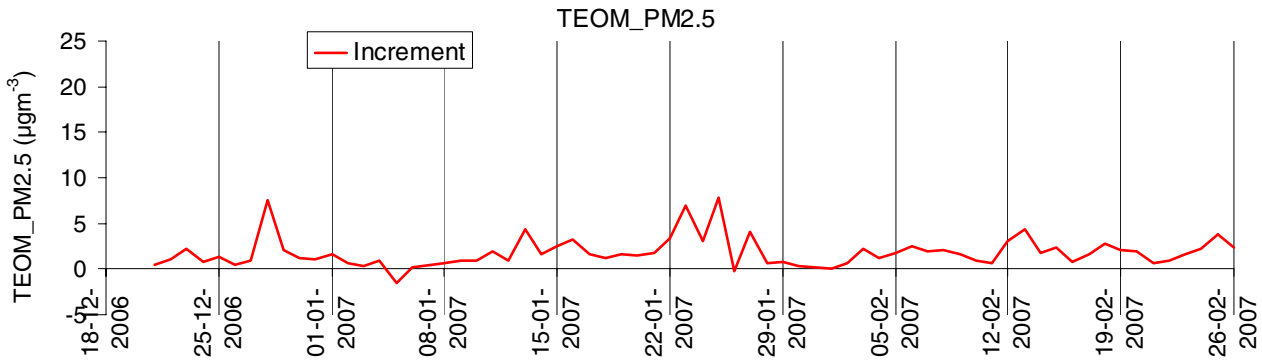


Figure 3.28 (a): Time series of concentration of $PM_{2.5}$ at the exposed (residential) site and at the background station. The values are TEOM measurements (uncorrected, and can therefore not be directly compared to the EU limit value). The curves are based on daily averages.
(b): Time series of the increment (the difference between the two curves).

3.6 Results for PAH

As described in section 2.3.5, 24-hour samples were collected during 10 non-consecutive days in order to measure PAH, levoglucosan and mannosan. The sampling began in the morning of the indicated day. During 8 of the 10 days simultaneous data were available for both the exposed site and the background site. Some main results for measured ambient concentrations of PAH are presented in Figure 3.29. The values concern only PAH in particulate phase.

A total of 15 PAH compounds were measured. Figure 3.30 shows the contribution of each PAH to the total PAH concentration, and the results are tabulated in Appendix A. Figure 3.31 shows results of measurements of monosaccharide anhydrides (also listed in Appendix A).

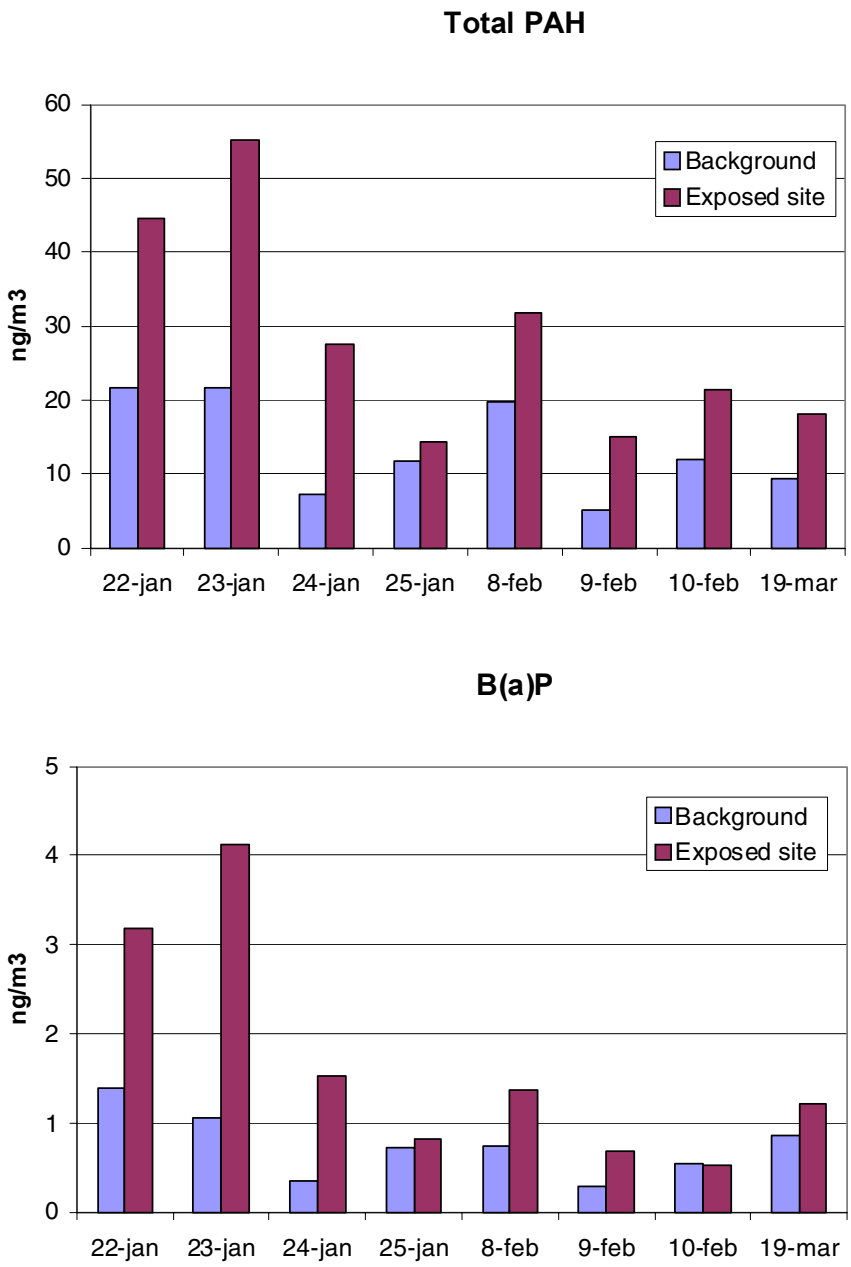


Figure 3.29. Results of measurements of total PAH (top) and B(a)PYR (or BaP, bottom), using 24h samples on quartz filter. The sampling began in the morning of the indicated day.

For the 8 days the average concentration of PAH at the exposed site was 28.5 ng/m^3, while at the background site it was 13.6 ng/m^3. Thus, PAH concentration at the exposed site was elevated by a factor of 2.1 compared to the background station. The average concentration of benz(a)pyrene (BaP) at the exposed site was 1.68 ng/m^3, while it was 0.74 ng/m^3 for the background site. This corresponds to an increment in BaP concentrations by a factor of 2.3. The value at the exposed site is somewhat higher than the target value for BaP on an annual average (1 ng/m^3) (Directive 2004/107/EC). Note, however, that the measured average values are based on only 8 days; thus, they are of limited representativity and do not represent a yearly average.

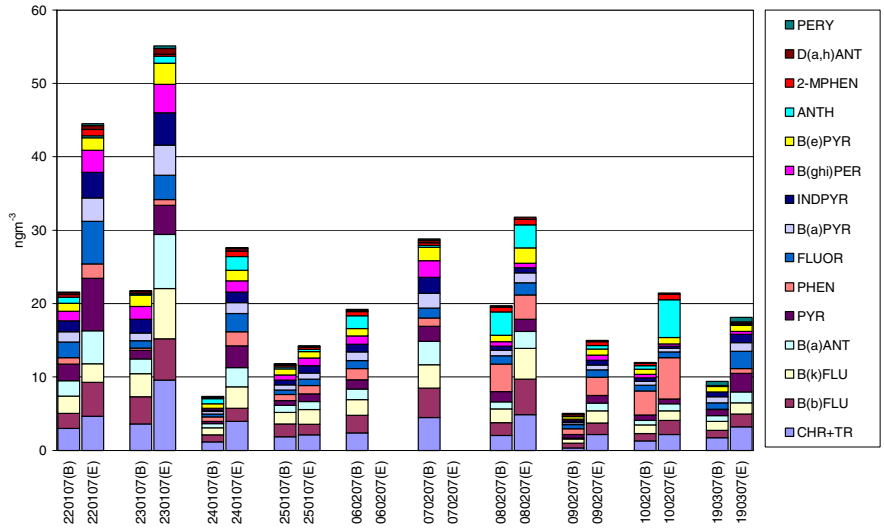


Figure 3.30. Corresponds to Figure 3.29, but gives the contribution of the single compound to the total concentration. The pairs of columns represent concentrations at the background site (B) and at the wood smoke exposed site (E).

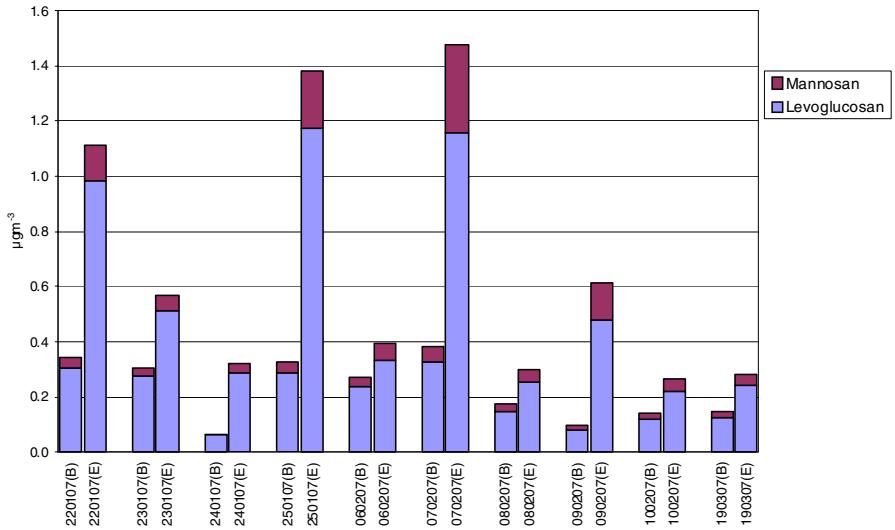


Figure 3.31. The ambient concentrations at the background site (B) and at the wood smoke exposed site (E) of mannosan and levoglucosan measured using 24h samples of particles on quartz filter.

3.7 Wood smoke source profile

3.7.1 $PM_{2.5}$, V, Soot, particle number N

Figure 3.25 shows the diurnal variation of the increments of $PM_{2.5}$, V and Soot for the cleaned data set. The fact that their variation is very similar indicates that one major source (the wood stoves) is the cause of the increments at the site in the village.

The curves for the modes of the N-increment in Figure 3.26 to some extent behave similarly to the curves of Figure 3.25, with most resemblance for the 65 nm mode and the 167 nm mode. However, the increments of N (especially the 13 nm mode and the 28 nm mode) seem to have some

contributions from the other local sources (traffic and the district heating station).

If another significant independent source of $PM_{2.5}$, V and Soot exists, it must have almost the same diurnal variation as the major wood stove source. We can test this by studying the co-variation of the time series of the 24 hour average values of the same variables during the entire campaign. This has been done using the COPREM receptor model (Wåhlin, 2003). Using a model (Eq. 1) with only one source

$$x_{ij} \cong a_i f_j \quad \text{(Eq. 1)},$$

where a_i is a constant 'wood smoke profile' for the different variables i, and f_j is the strength of the wood smoke source in sample j, we get a satisfactory prediction of measured value x_{ij} of the variables $PM_{2.5}$, V and Soot in the 24h samples as shown in Figure 3.32. The prediction of the particle number N is less satisfying, thus supporting the hypothesis that other local sources than wood smoke contribute to the particle number. The average contributions (a_i, if f_j is normalized to an average value of one) are shown in Table 3.2.

The value in the wood smoke profile for N may be too large because the average contributions from other sources are constrained to be zero in the one-source model. The position of the lower edge of the scattered points in Figure 3.32 indicates that the slope of the fitted line may be up to 50 % overestimated. On the other hand the strong variation of the shape of the size distribution shown in Figure 3.21, and especially in Figure 3.22 with data only from periods when the wood combustions source is dominating, indicates that a large part of the scatter is due to a strong variation of the number/mass ratio from wood combustion. A simple physical explanation can be hypothesised for the strong variation: When the mass concentration of the smoke is high, also the particle surface area is high leading to increased surface deposition of small particles with high mobility. As a consequence, the particle number concentration does not increase proportionally to particle mass concentration.

This analysis and the results in Table 3.2 give us some very central information of a general nature:

The values in the table link the three measures $PM_{2.5}$, V and soot to each other, so that from knowledge of the increment of one of these, one can estimate the value of the other two - provided that the source responsible for the increment is wood smoke from a residential area similar to the Danish village Slagslunde.

Particle number N does not have a close link to these three measures.

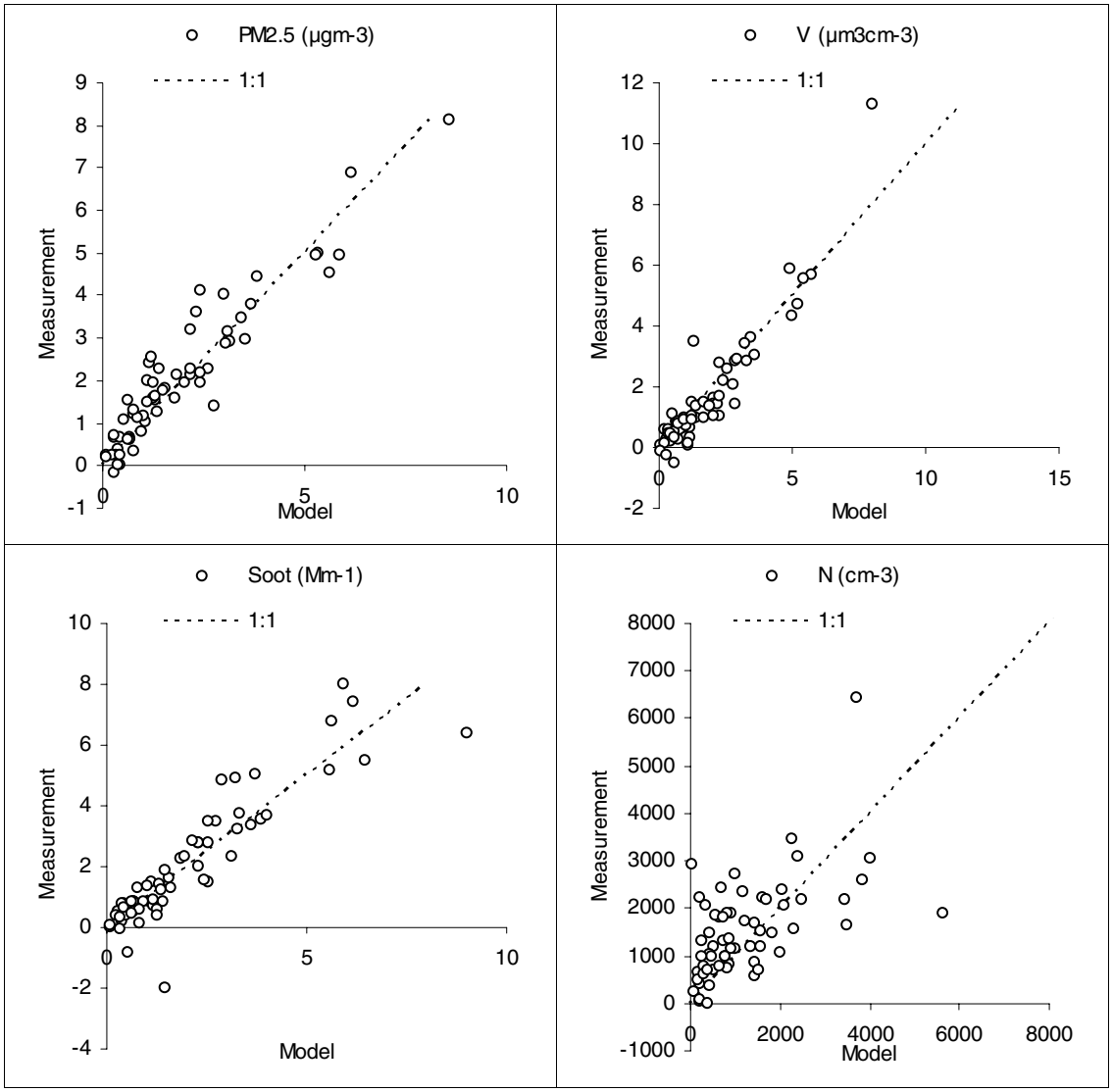


Figure 3.32. The COPREM fit compared with the measured values in 63 24h samples.

Table 3.2. The wood smoke source profile for $PM_{2.5}$, particle volume (V), light absorption (Soot), and particle number (N). The values are the average concentration increments in the village due to wood smoke using the cleaned data set for the entire campaign. The value for N may be up to 50 % overestimated, because other local sources (traffic and district heating station) probably have some influence on the particle number. The uncertainties are the standard deviations calculated within COPREM by unweighted linear regression analysis.

$PM_{2.5}$	1.91±0.05	μgm^{-3}
V	1.78±0.07	$\mu m^3 cm^{-3}$
Soot	2.01±0.08	Mm^{-1}
N	1253±154	cm^{-3}

3.7.2 PAH

The increments of the PAHs were calculated for seven 24h samples in which all the measured values at the background site and the exposed site were above the detection limit, and for which also data for $PM_{2.5}$, V and Soot were available. An unweighted regression analysis was made for each of the PAH increments as function of the source strengths found by the COPREM model (Eq. 1) using the average values of $PM_{2.5}$, V and Soot in each of the 24h PAH sampling periods. The correlation between the PAH increments and the $PM_{2.5}$, V and Soot increments were poor,

which is reflected in the high uncertainties on the PAH values in the resulting source profile (**Table 3.3**).

Table 3.3. Wood smoke source profile. The values in the table represent the averages of the increments during seven 24h PAH sampling periods.

$PM_{2.5}$	$PM_{2.5}$	2.9±0.2	μgm^{-3}
V	Particle volume	2.67±0.13	$\mu m^3 cm^{-3}$
Soot	Absorption	3.2±0.2	Mm^{-1}
CHR+TR	Chrysene + Triphenylene	1.7±0.7	ngm^{-3}
B(a)ANT	Benzo(a)anthracene	1.6±0.5	ngm^{-3}
PYR	Pyrene	1.6±0.5	ngm^{-3}
FLUOR	Fluoranthene	1.2±0.4	ngm^{-3}
B(k)FLU	Benzo(k)fluoranthene	1.0±0.5	ngm^{-3}
B(a)PYR	Benzo(a)pyrene	1.0±0.3	ngm^{-3}
INDPYR	Indeno(1,2,3-cd)pyrene	0.9±0.2	ngm^{-3}
B(b)FLU	Benzo(b)fluoranthene	0.9±0.5	ngm^{-3}
B(ghi)PER	Benzo(ghi)perylene	0.8±0.2	ngm^{-3}
PHEN	Phenanthrene	0.5±0.4	ngm^{-3}
B(e)PYR	Benzo(e)pyrene	0.5±0.2	ngm^{-3}
ANTH	Anthracene	0.3±0.6	ngm^{-3}
2-MPHEN	2-Methylphenanthrene	0.19±0.10	ngm^{-3}
D(ah)ANT	Dibenz(a,h)anthracene	0.17±0.05	ngm^{-3}
PERY	Perylene	0.08±0.02	ngm^{-3}
TotPAH	Total PAH	12±4	ngm^{-3}

3.7.3 Levoglucosan and mannosan

The increments of the monosaccharide anhydrides (levoglucosan and mannosan) could be calculated for nine 24h periods. An unweighted regression analysis was made for each of the monosaccharide anhydrides increments as function of the source strengths found by the COPREM model (Eq. 1) using the average values of $PM_{2.5}$, V and Soot in each of the 24h sampling periods. The prediction of mannosan is even less satisfying than the prediction of levoglucosan. Only the scatter plot for levoglucosan is shown in Figure 3.33. The high uncertainties for Levoglucosan and Mannosan in the source profile (Table 3.4.) are a consequence of the poor correlation with $PM_{2.5}$, V and soot.

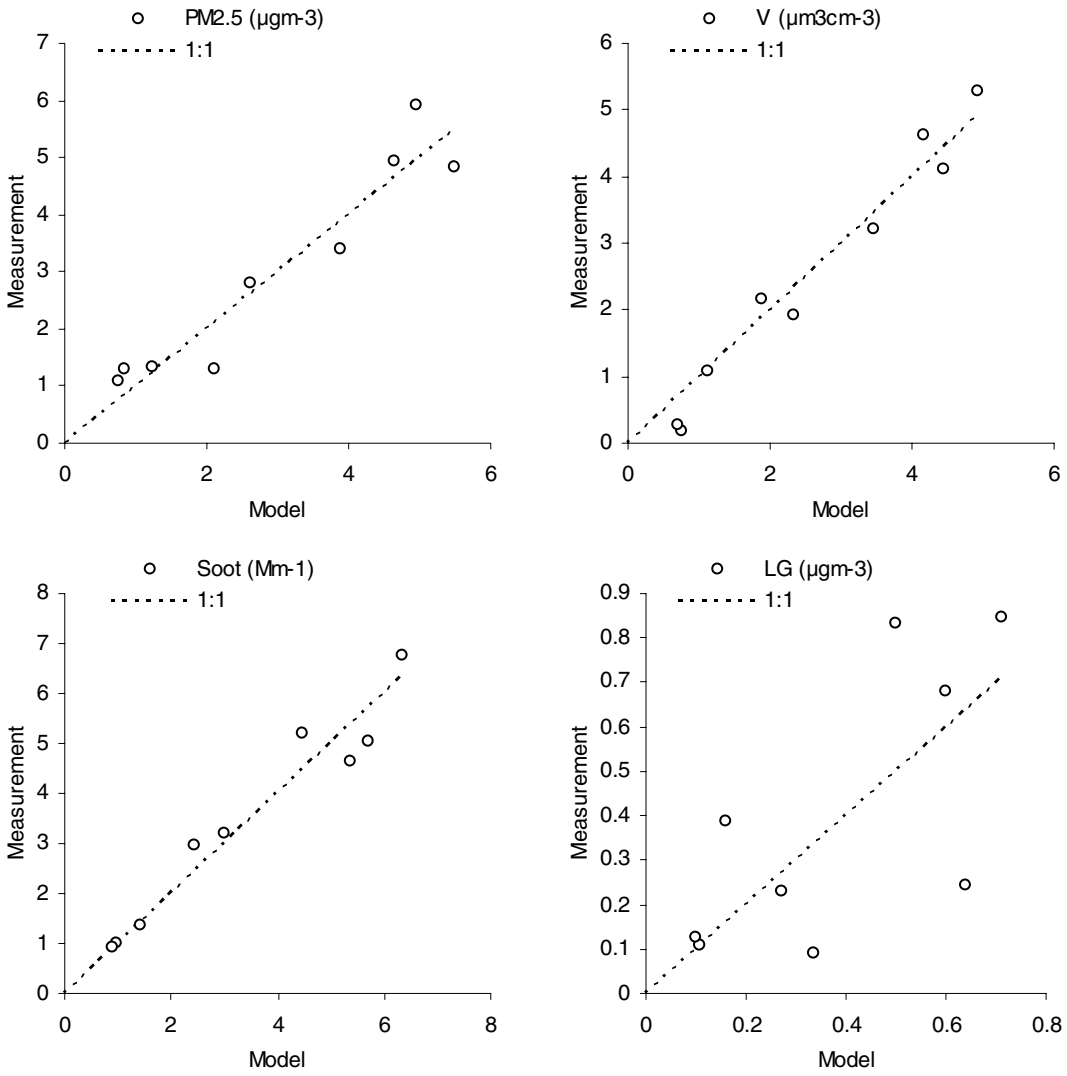


Figure 3.33. The COPREM fit of the increments of $PM_{2.5}$, particle volume (V), light absorption (Soot) and levoglucosan compared with the measured values in nine 24h samples.

Table 3.4. The values in the table represent the averages of the increments during nine 24h sampling periods of levoglucosan and mannosan.

$PM_{2.5}$	$PM_{2.5}$	3.0±0.2	μgm^{-3}
V	Particle volume	2.65±0.11	$\mu m^3 cm^{-3}$
Soot	Absorption	3.41±0.15	Mm^{-1}
LG	Levoglucosan	0.38±0.06	μgm^{-3}
Man	Mannosan	0.08±0.02	μgm^{-3}

4 Conclusion

$PM_{2.5}$, particle number size distribution, NO_x, CO, soot, PAHs and monosaccharide anhydrides were measured at two sites, one exposed by smoke from woodstoves in a small village (Slagslunde), and the other acting as background reference outside of the village. The increments (the differences in concentration between the exposed site and the background site) were calculated as measures for the local contributions. Meteorological measurements were performed from a mast placed on the top of a hill close to the background site.

Simultaneous measurements of NO_x were only available for 23 days. The purpose of the NO_x measurements was to verify that the traffic is not an important local source for the increment values of particle number and $PM_{2.5}$. This is indeed the conclusion for $PM_{2.5}$ for which the traffic contribution is estimated to be only ± 0.08 μgm^{-3} compared with the contribution from the wood stoves (1.9 μg^{-3}). However, for the *particle number concentration* a significant part (10-20 %) may be due to local traffic.

For further investigation of possible disturbing local particle sources, the *particle size distributions* turned out to be useful. For the sake of simplicity the increments of the detailed particle size distributions were replaced by the increments of four log-normal modes at 13 nm, 28 nm, 65 nm, and 167 nm, which gave the best overall fit. By studying the time series and the wind direction dependence of the individual modes, some episodes and wind directions were identified with contributions from other local sources than wood stoves. Most important were contributions to the 28 nm mode from local traffic in the wind sector 90°-130°, and contributions to the 13 nm mode from the district heating and power station. The influence on the 13 nm increment was positive when the wind came from the stack of the station to the site in the village, and the influence was negative when the wind was in the direction from the stack to the background site. Based on such analyses a cleaned data set was created in which all time periods with presumed influence from other sources than wood combustion was removed.

Curves for the average diurnal variation of the increments measured with high time resolution and based on the cleaned data set have almost the same shape for $PM_{2.5}$, for particle volume and for soot, thus providing strong evidence for the existence of only one major source, i.e. wood burning. Furthermore, the shape of the curves is typical for normal firing habits with low values during most of the day until the middle of the afternoon when residents return from work. At this point the values start to increase to a higher level and remain high in the evening until normal bedtime before or around midnight. The diurnal variation of the total *particle number* concentration is slightly different showing some increase already when the morning traffic starts. By an examination of the different modes it turns out this behaviour is mostly due to the smallest particles, represented by the 13 nm mode and the 28 nm mode. The behaviour of the 65 nm and 167 nm modes are very similar to the behaviour of $PM_{2.5}$, particle volume and soot. The conclusion is that the increments of $PM_{2.5}$, soot, particle volume and particle number in the larger size frac-

tions are almost entirely due to wood combustion, while some part of the particle number in the finer size range (28 nm mode) is due to traffic.

A central result of the study is a 'wood smoke source profile' - i.e. a vector which links the three measures $PM_{2.5}$, V and soot to each other, so that from knowledge of the increment of one of these, one can estimate the value of the other two - provided that the source responsible for the increment is wood smoke. Particle number N does not have a close link to these three measures.

It is an implication of the results that measures of particle number will not yield a fair impression of pollution from wood combustion. There is *some* correlation between particle number and other measures for wood smoke pollution, but particle numbers are not suitable as predictors for these other measures - particle number is a measure which is susceptible to contributions from other sources than wood smoke, and the results suggest that even for pure wood smoke there is no simple relation between particle numbers and particle mass. The calculated number in the average source profile may be up to 50% overestimated.

The PAH, levoglucosan and mannosan were measured in a limited number of 24 hour PM samples. All the other variables were measured with half-hour time resolution, but 24 hour synchronous averages were used for the calculation of wood stove combustion source profiles involving $PM_{2.5}$, soot, particle volume, particle number, PAH, levoglucosan and mannosan.

The level of PAH was clearly elevated at the exposed site compared to the background site. Data can be compared for 8 days only. For total PAH the average concentration was a factor of 2.1 higher at the exposed site as compared to the background site. For benz(a)pyrene (BaP) the concentration was increased by a factor of 2.3. The average BaP concentration at the exposed site for the very limited period of 8 days was 1.68 ng/m^3. The available data material is much too limited to allow any conclusions about the annual average, but for comparison it can be noted that the EC limit value for BaP is 1 ng/m^3 as an annual average.

5 Acknowledgements

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Appendix A

Table A.1 Atmospheric concentrations of PAH (ng/m^3). The column headers indicate an abbreviated name of a compound. The full names are given in Table A.3.

Date	Site	PHEN	ANTH	2-MPHEN	FLUOR	PYR	B(a)ANT	CHR+TR	B(b)FLU	B(k)FLU	B(e)PYR	B(a)PYR	PERY	INDPYR	D(a,h)ANT	B(ghi)PER	Tot PAH
22 Jan 2007	Backgr.	0.88	0.85	0.34	2.15	2.25	2.10	3.00	2.03	2.36	1.07	1.38	0.11	1.48	0.25	1.32	21.57
22 Jan 2007	Exposed	1.93	0.29	0.84	5.82	7.17	4.49	4.65	4.64	2.50	1.72	3.18	0.26	3.48	0.53	2.98	44.50
23 Jan 2007	Backgr.	0.30	0.18	0.10	1.02	1.18	1.99	3.59	3.70	3.17	1.54	1.06	0.09	1.83	0.23	1.76	21.74
23 Jan 2007	Exposed	0.77	0.96	0.27	3.33	3.99	7.33	9.59	5.63	6.86	2.90	4.12	0.35	4.38	0.77	3.88	55.10
24 Jan 2007	Backgr.	0.62	0.68	0.18	0.41	0.30	0.54	1.17	0.96	0.97	0.58	0.34	0.05	0.22	0.09	0.23	7.34
24 Jan 2007	Exposed	1.92	1.86	0.77	2.45	2.97	2.64	3.93	1.79	2.92	1.43	1.53	0.12	1.43	0.32	1.52	27.62
25 Jan 2007	Backgr.	0.83	0.27	0.20	0.60	0.67	0.93	1.88	1.73	1.58	0.86	0.72	0.06	0.63	0.13	0.68	11.78
25 Jan 2007	Exposed	1.12	0.31	0.25	0.89	1.03	1.12	2.11	1.43	1.99	0.89	0.82	0.07	0.99	0.16	1.03	14.24
06 Feb 2007	Backgr.	1.51	1.71	0.60	1.12	1.29	1.40	2.37	2.41	2.16	1.01	1.14	0.09	1.06	0.22	1.15	19.22
06 Feb 2007	Exposed				3.69	3.70	5.26	3.37									16.02
07 Feb 2007	Backgr.	1.14	0.26	0.33	1.34	2.04	3.19	4.48	4.01	3.17	1.83	2.05	0.17	2.18	0.35	2.26	28.80
07 Feb 2007	Exposed																0.00
08 Feb 2007	Backgr.	3.74	3.21	0.64	1.14	1.41	0.96	2.01	1.74	1.89	0.87	0.74	0.05	0.59	0.12	0.58	19.67
08 Feb 2007	Exposed	3.34	3.15	0.72	1.65	1.65	2.31	4.85	4.85	4.18	2.09	1.36	n.d.	0.67	0.31	0.64	31.78
09 Feb 2007	Backgr.	0.78	0.10	0.19	0.58	0.46	0.12	0.35	0.61	0.59	0.36	0.28	0.05	0.25	0.07	0.22	5.02
09 Feb 2007	Exposed	2.48	0.51	0.52	1.01	1.03	1.05	2.13	1.58	1.68	0.81	0.67	0.05	0.65	0.11	0.69	14.97
10 Feb 2007	Backgr.	3.25	0.45	0.33	0.80	0.74	0.63	1.28	1.01	1.16	0.68	0.55	0.05	0.46	0.10	0.51	12.00
10 Feb 2007	Exposed	5.65	5.15	0.75	0.76	0.65	0.93	2.15	1.92	1.34	0.88	0.51	n.d.	0.29	0.18	0.30	21.45
19 Mar 2007	Backgr.	0.04	0.02	0.02	0.88	0.81	0.78	1.71	1.01	1.23	0.77	0.85	0.55	0.63	0.08	n.d.	9.39
19 Mar 2007	Explosed	0.68	0.11	0.14	2.34	2.53	1.46	3.21	1.74	1.55	0.83	1.20	0.63	1.13	0.22	0.36	18.13

Notes: Missing values (blank fields) for some PAHs were due to missing recovery of the corresponding deuterium-labelled compounds. This might be due to a high concentration of organic carbon in the particles with consequent adsorprion of the compounds.

'n.d.' indicates a value below the detection limit

Table A.2. Levoglucosan and Mannosan

	Site	Levoglucosan µg/m^3	Mannosan µg/m^3
22 Jan 2007	Backgr.	0.30	0.04
22 Jan 2007	Exposed	0.98	0.13
23 Jan 2007	Backgr.	0.28	0.03
23 Jan 2007	Exposed	0.51	0.06
24 Jan 2007	Baggrund	0.06	0.00
24 Jan 2007	Exposed	0.29	0.03
25 Jan 2007	Backgr.	0.29	0.04
25 Jan 2007	Exposed	1.18	0.21
06 Feb 2007	Backgr.	0.24	0.03
06 Feb 2007	Exposed	0.33	0.06
07 Feb 2007	Backgr.	1.16	0.32
07 Feb 2007	Exposed	0.33	0.05
08 Feb 2007	Backgr.	0.15	0.03
08 Feb 2007	Exposed	0.25	0.05
09 Feb 2007	Baggrund	0.08	0.02
09 Feb 2007	Exposed	0.48	0.13
10 Feb 2007	Baggrund	0.12	0.02
10 Feb 2007	Exposed	0.22	0.04
19 Mar 2007	Baggrund	0.12	0.02
19 Mar 2007	Exposed	0.24	0.04

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