Joonas Enroth

Variation of Air Pollutant Concentrations in Residential Areas of the Helsinki Metropolitan Region

Helsinki Metropolia University of Applied Sciences
Bachelor of Engineering
Environmental Engineering
Thesis
28 May 2013
Residential wood combustion is one of the largest pollution sources in Finland. The purpose of this study was to look at the spatial and temporal variation of pollutants in the Helsinki metropolitan region. The primary interest was in observing the effects of residential wood combustion on the air quality and in studying how wood combustion emissions differ from traffic emissions.

The measurements were carried out during two weeks in February 2012 as a part of the MMEA (Measurement, Monitoring and Environmental Assessment) -project. The measurements were performed with Metropolia’s mobile air quality laboratory, Sniffer. The measurement campaign was realized in cooperation with the Finnish meteorological institute, Technical University of Tampere and Helsinki Region Environmental Services Authority.

During the campaign, 52 hours’ worth of data with one second time resolution were measured. The findings show that wood combustion creates a characteristic emission which is distinguishable from other sources. A period of long range transport of pollutants coincided with the measurement period, and it was found to be the dominating factor in air quality while present.

On the basis of the measurements, it seems that wood combustion has a significant effect on the air quality in the measured residential areas. The effects of wood combustion were found to be lower during the mornings than the evenings.

The most notable effects of wood combustion were found to be the particle emissions, where elevated levels of particles in the size range of 100-250 nm were observed. Traffic emissions were found to have their highest particle concentration in the smaller < 100 nm size range. The thermal treatment of samples showed wood combustion emissions having low volatility, while traffic emissions typically were found to be highly volatile.
Contents

1 Introduction 1

2 Ambient air 2
   2.1 Particulate matter 3
   2.2 Traffic emissions 5
   2.3 Wood combustion 5

3 Experimental 6
   3.1 Measurement campaign 6
   3.2 Objectives 7
   3.3 Metropolia Mobile Air Quality Lab - Sniffer 7
      3.3.1 Instrumentation 9
      3.3.2 Measurement Set up 13
   3.4 Residential Wood Combustion Areas 15
   3.5 Data analysis and Quality assurance 16

4 Results and Discussion 17
   4.1 Weather 20
   4.2 Long-range transport of pollution 20
   4.3 Particle Measurements 22
   4.4 Gaseous pollutants 26
   4.5 Urban background measurement 27
   4.6 Residential areas 27
      4.6.1 Source attribution 29
      4.6.2 Temporal distribution 31
      4.6.3 Spatial distribution within the residential areas 32

5 Conclusions 37

References 40

Appendices

Appendix 1: Example of RWC and Traffic Emissions
Appendix 2: Effects of temporal variation on air quality
Appendix 3: Temperature during the measurements
Appendix 4: Mean pollutant concentrations measured in the residential areas
Appendix 5: Measurement results by area
**Abbreviations**

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaP</td>
<td>benzo(a)pyrene</td>
</tr>
<tr>
<td>CLEEN</td>
<td>Cluster for Energy and Environment</td>
</tr>
<tr>
<td>EPA</td>
<td>U.S. Environmental Protection Agency</td>
</tr>
<tr>
<td>FMI</td>
<td>Finnish Meteorological Institute</td>
</tr>
<tr>
<td>HSY</td>
<td>Helsinki Region Environmental Services Authority</td>
</tr>
<tr>
<td>LRT</td>
<td>Long Range Transport</td>
</tr>
<tr>
<td>MMEA</td>
<td>Measurement, Monitoring and Environmental Efficiency Assessment</td>
</tr>
<tr>
<td>NDIR</td>
<td>Non Dispersive Infra-Red</td>
</tr>
<tr>
<td>PM</td>
<td>Particulate Matter</td>
</tr>
<tr>
<td>PMF</td>
<td>Positive Matrix Factorization</td>
</tr>
<tr>
<td>RWC</td>
<td>Residential Wood Combustion</td>
</tr>
<tr>
<td>TUT</td>
<td>Technical University of Tampere</td>
</tr>
</tbody>
</table>
1 Introduction

Air pollution is a topic of growing interest. Air pollution is defined by the Encyclopedia Britannica (2012) as: “Release into the atmosphere of various gases, finely divided solids, or finely dispersed liquid aerosols at rates that exceed the natural capacity of the environment to dissipate and dilute or absorb them”. Natural sources produce air pollution as the conspicuous omission of human interference from the definition implies. One just has to think about forest fires or volcano eruptions, and it is easy to see that these also produce air pollution. Other natural sources include dust storms and sea spray. Air pollution thus has been around much longer than humans have.

Anthropogenic air pollution is air pollution that is caused by human activity. Already in the 14th century London there was legislation banning coal burning while the parliament was in session, due to the pollution it created. Since then air pollution has been thought of as a nuisance, and only in recent times it has been understood that air pollution can have more serious effects. The mid-20th century saw some of the all-time strongest smog episodes in London and Los Angeles and raised the awareness of the importance of air pollution control. By the 1980’s air pollution was generally recognized to have serious effects on the global scale with the discovery of ozone depletion and greenhouse gases. (Vallero, 2008, p. 40-49)

Since then topics like acid rain and global warming have been introduced to the public, and a greater understanding of the potential harms associated with air pollution have slowly been understood. One of these more recently recognized forms of air pollution is particulate matter (PM), which, in brevity, is still largely not understood outside the scientific community. (De Nevers, 2000; Vallero, 2008)

The purpose of this thesis was to look at the ambient air quality of residential areas in the Helsinki metropolitan region. The main focus of the thesis was in studying the effects that residential wood combustion has on the air quality and how do the traffic emissions differ from wood combustion emissions.
2 Ambient air

Ambient air is the air that we breathe every day. This constant exposure makes the study of ambient air an important topic. Table 1 below gives the limit values that are in use in Finland for ambient air.

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Averaging Time</th>
<th>Limit (293 K, 101,3 kPa)</th>
<th>Annual allowed exceedences</th>
<th>In effect since</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sulfur Dioxide (SO₂)</td>
<td>1 hour</td>
<td>350 µg/m³</td>
<td>24</td>
<td>1.1.2005</td>
</tr>
<tr>
<td></td>
<td>24 hours</td>
<td>125 µg/m³</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nitrogen Dioxide (NO₂)</td>
<td>1 hour</td>
<td>200 µg/m³</td>
<td>18</td>
<td>1.1.2010</td>
</tr>
<tr>
<td></td>
<td>1 year</td>
<td>40 µg/m³</td>
<td>-</td>
<td>1.1.2010</td>
</tr>
<tr>
<td>Particle pollution (PM₁₀)</td>
<td>24 hours</td>
<td>50 µg/m³</td>
<td>35</td>
<td>1.1.2005</td>
</tr>
<tr>
<td></td>
<td>1 year</td>
<td>40 µg/m³</td>
<td>-</td>
<td>1.1.2005</td>
</tr>
<tr>
<td>Lead</td>
<td>1 year</td>
<td>0.5 µg/m³</td>
<td>-</td>
<td>15.8.2001</td>
</tr>
<tr>
<td>Particle pollution(PM₂.₅)</td>
<td>1 year</td>
<td>25 µg/m³</td>
<td>-</td>
<td>1.1.2010</td>
</tr>
<tr>
<td>Carbon Monoxide(CO)</td>
<td>8 hours</td>
<td>10 mg/m³</td>
<td>-</td>
<td>1.1.2005</td>
</tr>
<tr>
<td>Benzene (C₆H₆)</td>
<td>1 year</td>
<td>5 µg/m³</td>
<td>-</td>
<td>1.1.2010</td>
</tr>
</tbody>
</table>

In the last decades great advances have been made in improving the air quality. The levels of SO₂, CO and NOx have gone down significantly. While progress has been made in combating these gaseous pollutants, the situation with particulate matter has not been significantly improved. (Finnish Environment Institute, 2013)

Weather is one of the key factors that determine ambient air quality. Weather controls the movement of long range transport and the formation of inversion layers, both being factors that can significantly impact the air quality of a given area. Windy conditions promote atmospheric mixing, that results in cleaner air by diluting the pollutants over larger area. Rain and cloud formation usually clean the air by capturing pollutants to the raindrops and removing them from the air either by washout or rainout. Cold weather requires more heating, which usually means more combustion of fossil fuels and thus leading to increased emission rates.

Most of anthropogenic air pollution results from combustion of fossil fuels, contributing to the CO₂ content of the atmosphere. In practice, combustion always also produces a wide array of other pollutants besides CO₂ due to incomplete combustion or impurities
in the fuel. Some of the most common ones include VOCs, CO, SO₂, NO₂, lead and particulate matter (Vallero, 2008).

2.1 Particulate matter

Particulate matter (PM) is a term used to describe small particles that are suspended in the air. They can be either liquid droplets or solids. Their classification is based entirely on the particle size. Common classifications are PM₁₀, PM₂.₅ and PM₁ meaning particles with a diameter less than 10, 2.5 and 1 μm, respectively (EPA, 2013). The negative health effects of these particles make them a cause of concern. According to the Helsinki Region Environmental Service Authority (HSY), the most harmful air pollutant for human health in the Helsinki metropolitan region is particulate matter (Malkki, et al., 2012, p.8).

In addition, particles interact with the climate through direct and indirect effects. Direct effects include for example scattering, and in the case of soot particles, absorption of solar radiation. Indirectly they can have an effect by acting as cloud condensation nuclei. (Pirjola et al., 2012; Seinfeld and Pandis, 2006)

Particulate matter affects humans through the respiratory system. PM₁₀, or thoracic particles, can penetrate deep into the respiratory system, the PM₂.₅, or fine particles, can enter even deeper in to the alveoli and the ultrafine particles(< 0.1 μm) can penetrate all the way through the lungs and enter the bloodstream. Elevated levels of particles are known to cause an increase in hospital admissions, and a rise in mortality. (Malkki, et al., 2012, p. 9)

It is commonly understood that particulate matter can have serious adverse health effects. Most of the effects impact the respiratory system, but they can also affect the entire body. The inhaled particulates cause inflammation and can act as a trigger to various medical conditions. More serious tough are the effects of chronic exposure to elevated levels of particulate matter. This causes the body to be chronically in a condition of mild stress, thus enabling the formation of many medical conditions, slowing recovery rates, and even causing premature deaths. (Salonen and Pennanen, 2006)

Figure 1 depicts the typical modes of number and volume distributions of atmospheric particles. It shows how the ultrafine region (< 0.1 μm) is mostly made up of particles in
the nucleation and Aitken mode. The nucleation mode is formed via the gas-to-particle conversion that results in the formation of nuclei. The Aitken mode consists largely of primary particles. Primary particles are particles that have been directly emitted in particle form, whereas secondary particles are particles that form in the atmosphere. (Seinfeld and Pandis, 2006)

Particles with a diameter less than 2.5 μm are known collectively as fine particles, while the particles larger than 2.5 μm are called coarse particles. Fine particles in the accumulation mode are mostly secondary particles that have grown in the atmosphere from ultrafine particles. Most coarse particles form as the result of mechanical processes and thus are primary particles. (Seinfeld and Pandis, 2006)

Figure 1. Typical particle number and volume distributions of particles in the atmosphere. (Seinfeld and Pandis, 2006)
As can be seen in Figure 1, the smaller particles have the largest impact in the number range, while having virtually no mass. This is due to their extremely small size, resulting in an extremely small volume. The smaller particles also have a large surface area, and are prone to adsorb various compounds or metals on their surface. The larger particles come in much smaller numbers, but as they can be hundred or thousand times larger they dominate the mass distribution.

In urban areas most of the PM originates from traffic or energy production. Traffic produces PM not only via combustion but also in the form street dust, which can contribute significant amounts to the total, especially during spring time. (Malkki, et al., 2012) Not all particulate matter sources are anthropogenic, and nature produces PM in such forms as sea spray, desert dust or combustion emissions through forest fires. (Vallero, 2008, p. 59)

In the atmosphere particulate matter has a lifetime ranging from some minutes to weeks and it is removed either by dry or wet deposition. Dry deposition usually means methods such as impaction, interception, diffusion, turbulence and gravity settling. Wet methods include scavenging through precipitation or snowfall and the through the effects of fog and clouds. (Seinfeld and Pandis, 2006)

2.2 Traffic emissions

The largest source of pollutants in the Helsinki metropolitan region is traffic. It produces up to 32 % of NOx, 45 % of particles and 81 % of CO generated in the region. Virtually all of the particles resulting from traffic are generated by diesel vehicles, while more than 85 % of CO emissions are from gasoline vehicles. (Malkki et al., 2012) In addition to the engine emissions, traffic produces road dust that also affects the urban air quality. This effect is heightened during the winter and especially spring season when studded tires are used.

2.3 Wood combustion

In suburban areas where there is less traffic, the importance of wood combustion as a particle source is elevated. According to a recent study, up to 60 % of PM in suburban areas can originate from residential wood combustion (RWC). (Saarnio et al., 2012)
Although only 2 per cent of the houses in the Helsinki metropolitan area have wood as their primary source of heat, a total of 80% of detached houses have wood stoves, sauna or use wood for auxiliary heating. It estimated that one sixth of all PM emissions in the Helsinki metropolitan area, result from wood combustion. (HSY, 2013a)

Although modern combustion technologies can reduce the generation PM significantly, most of the combustion appliances used in Finland are still conventional batch burning installations and therefore likely to have a recognizable effect on the air quality of areas that have dense wood combustion (Lamberg et al., 2011).

Small scale wood combustion is very heterogenic source of emissions. Factors such as fuel moisture and type of the wood, as well as the type of the stove, can affect the resulting pollutants heavily (HSY, 2013a). In clean combustion, where only dry and untreated wood is being burned, the emissions are much smaller and produce less particulate matter. These variations of different combustion methods, locations and events have been previously studied in laboratory environment, but the study of particles in ambient air has been limited. (Weimer et al., 2008; Lamberg, et al., 2011)

3 Experimental

3.1 Measurement campaign

The measurements campaign reported in this thesis was part of the Measurement, Monitoring and Environmental Efficiency Assessment (MMEA) -program that is funded by the National Technology Agency (TEKES) and private companies. The project is coordinated by Cluster for Energy and Environment (CLEEN). The project was a part of work package 4.5.2, whose coordinator is Finnish Meteorological Institute, and other participants included Technical University of Tampere, Metropolia UAS and HSY.

The measurements took place in the period spanning from 15th to the 27th of February 2012. All measurements were performed with the Metropolia mobile air quality laboratory, Sniffer, and concentrated on six residential areas located in the Helsinki metropolitan region. Three of the areas were located to the west of Helsinki, in Espoo, and three were to the east in Vantaa and Helsinki. The measurements were performed on alternating days in the west and in the east. In addition to the residential areas, the air quali-
ty of the city center was measured before each route and an urban background after each route.

Wood burning usually occurs during two peaks, one in the morning and one in the evening (Myllynen, 2010). The measurements were mostly conducted during the evening peak as it is the more prominent of the two. A total of 12 measurement runs were made, of which two were done in the morning from 9 to 12, on the 21st and 24th of February, and the rest in the evening from 17 to 23. No measurements were made on the 19th, 22nd or the 23rd.

3.2 Objectives

The objectives for the measurement campaign reported in this thesis included the determination of the physical and chemical characterization of PM produced by RWC and traffic emission, as well as their effects on the air quality of suburban areas in the Helsinki region. Additional points of interest included studying the spatial variation of these pollutants and to complimenting the stationary measurements conducted in the same residential areas simultaneously by HSY. (HSY, 2011)

Another key interest was to record the variations in air quality between the different measurement areas, as well as to observe the temporal variation in air quality. Particle concentrations as well as the physical properties of the particles resulting from both RWC and traffic were to be measured so as to enable the determination of the proportion of particles resulting from RWC and traffic in these areas.

An additional motivation for this measurement campaign was to gain more data on the effects of RWC in situ. As stated before, the amount of data on the effects on wood combustion in real life situations is limited compared to laboratory data. The focus here was to look at the variation of pollutants with very high spatial resolution. Thus far only few studies have looked at the variation of pollutants with as high spatial resolution as is done here. (Weimer et al., 2008)

3.3 Metropolia Mobile Air Quality Lab - Sniffer

The Metropolia Air Quality Research unit is centered on the Sniffer vehicle, a mobile laboratory that was built in 2003 as a part of air quality research program LIPIKA (Pirjo-
la et al., 2004). Sniffer is a fully functioning air quality measurement laboratory built into a Volkswagen LT35 diesel van, shown below in Figure 2. It has a length of 5585 mm, a width of 1933 mm, and height of 2570 mm. Sniffer was built with road vehicle emissions measurement in mind, but it has proved to be a versatile platform for other air pollutant measurements as well.

Figure 2. The Sniffer vehicle. The air inlet lines used for the measurements can be seen above the windshield. (Metropolia, 2008)

Mobile air pollution measurement vehicles have unique measuring opportunities as they can capture the spatial attributes of air pollution often much more effectively than stationary measurement sites. In this work Sniffer measured and recorded particle size distributions and number concentrations ranging from 0.007 to 5 μm; PM₁ and PM₂.₅ mass concentrations; nitrogen oxides such as NO, NO₂, NOₓ; weather conditions such as temperature, relative humidity, wind speed and direction. Sniffer is also equipped to measure GPS-location, as well as CO and CO₂ concentrations.

For the duration of the measurement campaign Sniffer was further equipped with measurement devices from the Tampere University of Technology and Finnish Meteor-
ological Institute. Data from these devices were not available for this thesis and the results are not considered here.

3.3.1 Instrumentation

3.3.1.1 Elpi

Electrical Low Pressure Impactor (Elpi) by Dekati Ltd is a cascade type impactor device, used to measure particle concentrations and size distribution. The operating principle is one where the air sample is sucked through the device with a vacuum pump, as shown in Figure 3 below. The air sample is passed by a corona charger that charges the passing particles to a known charge by creating an electric field. (Dekati, n.d.)

![Figure 3. Working principle of the ELPI. (Dekati, n.d.)](image)

These charged particles are then passed through a cascade of sieves and collector plates, called the impactor. The impactor consists of a stack of twelve sieves with a collector plate placed under each sieve. All collector plates are connected with their individual electrometers. Due to the different hole size in the sieves, each stage has a
different separation capability, and thus the particles can be collected according to their respective sizes. (Dekati, n.d.)

When a charged particle hits a collector plate placed under a sieve, it sticks there and releases its charge. The current measured by the electrometer from each stage is directly proportional to the number of particles impacting that stage. (Dekati, n.d.)

Two Elpis were used for this measurement campaign, both of which are customized models that have a lower detection limit of 7 nm and the upper around 5 μm, with 12 stages. The other Elpi was additionally fitted with an extra stage in order to achieve higher resolution in the ultrafine particle size region. (Marjamäki and Keskinen, 2001; Yli-Ojanperä et al., 2010)

Thermal Denuder (TD) was used in front of one of the Elpis, the one without the fitted extra stage. Its purpose was to heat the sample air to around 265 °C, vaporizing all the volatile compounds in the sample. Some losses occur due the TD, but these losses have been taken into account and numbers have been corrected. (Rönkkö, 2011)

3.3.1.2 Teom 1400 ab

Teom, or the Tapered Element Oscillating Microbalance, is a particle mass concentration measurement device manufactured by Thermo Scientific. It is approved for PM$_{10}$ and PM$_{2.5}$ measurements by the U.S. EPA, but it can be used to measure also PM$_{1}$. (Thermo Scientific, 2004)

The operating principle is based on an oscillating microbalance where the particles are collected. It is by the minute changes in the oscillation frequency that the TEOM detects particles and their masses. TEOM has a total sample flow of 16.7 lpm, of which 13.7 lpm is used as auxiliary flow. The remaining 3 lpm is the actual sample which is heated to 50 °C in order to rid the air from excess moisture, which might affect the results. (Thermo Scientific, 2004)

The 3 lpm sample flow is directed to a mass transducer whereby it goes through a glass fiber filter. This filter is the oscillatory part which is sampled every two seconds, and the mass determined through the changes in the oscillations. After the mass has
been determined, the device converts the data to a mass concentration by using the known sample flow rate. (Thermo Scientific, 2004)

### 3.3.1.3 DustTrak II 8530

DustTrak II is a dust concentration measurement device manufactured by TSI. The device has an optical measurement system whereby the air sample is passed through a laser beam. The device measures the scattering of the beam, caused by particles in the laser’s path by a photometer. On the basis of the attributes of the diffraction, the device calculates the mass concentration of particles in the sample stream. (TSI, 2008)

The DustTrak II can detect aerosols, such as dust, smoke and mist. It can be used to measure either PM$_1$, PM$_{2.5}$ or PM$_{10}$ concentrations, depending on the particle size cut-off installed, and it has a detection range of 0.001 to 400 mg/m$^3$. (TSI, 2008)

Sniffer was fitted with two DustTraks, one measuring PM$_1$ and the other measuring PM$_{2.5}$. It should be noted that both devices have been calibrated with Arizona Test Dust.

### 3.3.1.4 VA-3000 – CO$_2$ analyzer

The VA-3000 is a gas analyzer manufactured by HORIBA used for measuring CO$_2$. It utilizes a non-dispersive infrared sensor, NDIR, to measure the CO$_2$ concentration. This is done by using a specific wavelength that is absorbed virtually only by CO$_2$, and detecting the absorption rate via a photometer. (Horiba, n.d.)

### 3.3.1.5 APNA-360

The APNA-360 is a Nitrogen Oxide Analyzer manufactured by Horiba. It measures the NO, NO$_2$ and NO$_x$ concentration by chemiluminescence (Horiba, 1997). The NO is measured by exposing the sample to O$_3$ which converts NO to NO$_2$. A fixed portion of the converted NO$_2$ will be at an exited state and will release light when returning to the ground state by itself, as described by the reaction below:

\[
NO + O_3 \rightarrow NO_2 + O_2 + hv
\]


The NO\textsubscript{x} concentration is calculated by converting all nitrogen oxides to NO, and then measuring the total concentration by the same method as described above. NO\textsubscript{2} is simply then calculated as the difference between NO and NO\textsubscript{x}. (Horiba, 1997)

### 3.3.1.6 CO12M

CO12M is a carbon monoxide analyzer by Environnement S.A designed to measure the low CO concentration that prevail in normal atmospheric conditions. It uses NDIR to measure the CO concentration. Carbon monoxide is highly effective at absorbing infrared radiation and its absorption maximum lies at 4.6 μm. The infrared used is also the same wavelength, in order to minimize the absorption to other materials. This is further ensured by the use of optical filter that eliminates the interference from other gases with similar spectra to that of CO. (Environnement S.A., n.d.)

The device has a time resolution of around two seconds and a lower detection limit of 50 ppb. Operational range is from 0.05 ppm to 20 ppm (Environnement S.A., n.d.).

### 3.3.1.7 Vaisala Weather Station

WS425 Ultrasonic Wind Sensor is a wind sensor manufactured by Vaisala. It measures wind speed by sending a signal from each of the three ultrasonic transducers to the two others, and calculating the difference in transition times. The waves are slightly slowed down by moving upwind, resulting in small differences in the transition times. From these differences the device can calculate both wind speed and direction. (Vaisala 2003)

HMP45A Temperature and Relative Humidity sensor measures relative humidity by a thin film polymer sensor, Humicap180, which changes its capacitance when it absorbs water molecules. The temperature is measured by resistive platinum sensors. Both of these sensors are cased in a shelter from direct exposure to the sun and rain. The casing was located on the roof of Sniffer at an altitude of 2.9 meters. (Vaisala, 1997)

### 3.3.1.8 Other measurement devices

For the duration of the measurement campaign also other devices were fitted in Sniffer. The most significant of these was the Aerosol Mass Spectrometer, or AMS, as it has
the capability to detect Levoglucon, a tracer compound resulting from wood combustion.

The Aethalometer is a measurement device for measuring black carbon (BC) concentration in air samples (Allen, 2006).

The Scanning Mobility Particle Sizer, SMPS, is a device for recording high resolution particle size distributions (Nanocap, ca. 2008).

The Pegasor Partice Sensor, PPS, is continuously operating particle mass concentration measurement device (Tikkanen, 2010).

The Nanoparticle Surface Area Monitor or NSAM is used to measure particle deposition area of nanoparticles in the human lungs (Nanocap, ca. 2008).

The devices described in this subsection were all controlled by the external parties, and are not considered in this report.

3.3.2 Measurement Set up

The sample air for the devices was taken from two separate inlets, located above the vans windshield at a height of 2.4 m. The smaller pipe with an inner diameter if 8 mm was used for the AMS and Aethalometer and the larger with and inner diameter of 26 mm was used for all other measurement devices, as shown below in Figure 4.
The DustTrak in the same sample line with the TEOM is distinguished as DustTrak B (DTB), from the DustTrak A in the separate line, referred to as DTA. DustTrak B and TEOM were operated mostly behind a cyclone with a cutoff at 2.5 μm, and DustTrak A, Aerosol Mass Spectrometer and the Aethalometer were operated behind particle size cutters with a limit a 1 μm.

During the first four measurement days from the 15th to the 18th, a cyclone with a cutoff of 1 μm was used for the sample line that provided the air to the DTB and TEOM. This cyclone was changed on the 20th of February, and the new cyclone was in operation already on that day. This configuration was used for the rest of the campaign. The new cyclone had a cutoff of 2.5 μm, and at the same time the cutoff for the DTA was changed to 1 μm, enabling the measurement of both PM$_1$ and PM$_{2.5}$ during all times of the campaign.

This change would have been visible in the TEOM data, and it might have had some effect on the results of the DustTraks (DTA and DTB) if the data were included here. The four first days coincided with a heavy period of long range transport (LRT), rendering the data useless and, therefore excluded from the thesis. Thus, the change did not have an effect on the results although the data from these four first days are still shown in Figure 6 and Appendix 5.
3.4 Residential Wood Combustion Areas

The areas that were chosen for the measurement sites represented typical suburban residential areas in the Helsinki metropolitan region. The chosen residential areas all have high percentage of houses with wood stoves and high occurrence of wood combustion, making these areas of interest. All of the measurement locations of the western route were situated in the city of Espoo, at Kattilalaakso, Laaksolahti and Lintuvaara. The eastern locations were situated in Vantaa and Helsinki, at Vartiokylä, Itä-Hakkila and Päiväkumpu. These locations, along with the background measurement site, are shown in Figure 5.

![Figure 5. Map of the measurement locations and urban background site. (OSM, 2013)](image)

The house composition of these areas was typical Finnish suburban composition, with the majority of the houses being detached houses, and to a lesser extent semi-detached houses and row-houses. None of the measurement areas has highways in their immediate vicinity, tough none of them is further than 2 km from a major highway either.
After each measurement route, the urban background was also measured in order to remove the effects of daily variation and long range transport interfering with interpretation of the results. The chosen location for the urban background measurement site, shown in Figure 5, was situated in Hietaniemi. The location is surrounded by the sea, a park and a cemetery. No major roads are in the immediate vicinity. Länsiväylä, a major highway, is located 550 meters to the south across a bay. Mechelininkatu, a busy road in the city is located 900 meters to the east, with a park and housing in between the road and the measurement site.

3.5 Data analysis and Quality assurance

In pollution monitoring, the data amounts are usually very large. Measurements typically span from a minimum of months to years or even decades. The Sniffer vehicle has a very different way of measuring air pollution. It is best suited to perform rapid measurement with spatial aspects involved. This also means that inevitably the data sets are smaller than usually encountered in the field, even though the time resolution is significantly higher in this method. In mobile measurement time resolution of one to two seconds is used, whereas stationary monitoring usually has a resolution ranging from minutes to hours. This is apparent when one looks how most limit values are expressed. Typically the limit values are given in yearly, monthly or sometimes daily exposures, and only very rarely hourly exposures are mentioned. (Malkki et al., 2012, p.14; HSY 2013b)

The interpretation of this limited amount of data in a scientifically valid way without over- or underestimating the results is one of the biggest challenges in this research method. This became especially important when the long range transport episode experienced at the beginning of the measurements rendered much of the data unusable. The amount of data left, after excluding the LRT portion, was quite small. The validity of the measurements was ensured by cross referencing them to the data provided by the HSY from their stationary measurement sites in the same areas.

The validity of the data had a high priority while conducting the measurements and analyzing the results. Steps that were taken to ensure the quality of the data included a zero point check done on all particle measurement devices before and after each measurement session. This was done by sampling air through a HEPA filter, cleaning
the air of any particulate matter. The gas analyzers have their internally built zero-point checks that were used daily.

There had been heavy snowfall before the measurement period, causing a significant build-up of snow on the roads. As the measurements were done in residential areas and on small roads, the snow caused some problems with the measurements. The built-up snow made the road surface extremely uneven, resulting in virtually constant vibrational stress on the measurement devices. This was sure to have an effect on the measured data. Especially the data from the TEOM seems to have suffered from the shaking, as the vibrations must have interfered with the oscillation of the microbalance. The data from the TEOM is not considered here, due to its suboptimal quality.

While conducting the measurements in the RWC areas all vehicle encounters were marked along with a time stamp into a log book, so as to account for the possible external disturbances in the data. Also all zero point drifts were recorded in the log book and referenced when analyzing the data.

4 Results and Discussion

The two week measurement period span from 15\textsuperscript{th} to the 27\textsuperscript{th} of February 2012. During this period a total of 12 successful measurement loops were made, consisting of 54 hours of data with one second time resolution. On the 21\textsuperscript{st} and the 24\textsuperscript{th} also morning measurements were performed from 9 to 12, while all other measurements were evening measurements and recorded between 17 and 23. Out of these twelve measurement days, the first four were affected by the presence of long-range transport (LRT), and thus only the period from 20\textsuperscript{th} to the 27\textsuperscript{th} day is considered here.

The effect of the LRT is highly visible from Figure 6 giving an overview of the measurement results. The figure presents all of the measurement data recorded within the residential areas, with the white columns indicating areas in the east and the grey areas in the west. Plots for the background, individual residential areas, and the city measurements can be found in Appendix 5.
Figure 6. Measurement results from the residential areas. White background is for eastern areas and the grey for the western areas. The black bar indicates median, the top and bottom of the box are the upper and lower quartiles and the whiskers denote the maximum and minimum values (1.5 times the corresponding quartile). Outliers are removed from the plot in order to improve clarity.

The presence of LRT is especially clear in the measured PM, where the concentrations were almost an order of magnitude higher during the LRT episode than otherwise. When comparing these values to the limit values provided in Table 1, one can see that the PM$_{2.5}$ concentrations were also significantly above the annual limit value set for PM$_{2.5}$ during the LRT episode.
Other noteworthy points are the lower morning concentrations on the first route of the 21st and the 24th, as well as the high concentrations of the Saturday evening on the 25th. The high concentrations on the 25th and on the 27th might partially be from slight presence of LRT. It is also worth noting the relatively high PM_{1} and PM_{2.5} concentrations found on Friday the 24th, as then there was no LRT at all present. This elevation is most probably the direct result of higher levels of wood combustion.

As can be seen from Figure 6, the daily variation was quite large with most pollutants. CO_{2} and CO had quite stable levels overall. NO had a very stable median, but the variability was quite large, possibly due to traffic.

In order to study the effects of residential wood combustion (RWC), the emissions resulting from wood combustion needed to be separated from other sources. This is traditionally done by utilizing characteristic compounds, usually known as tracers or markers, which act as an indicator of the particles’ origin. The markers used in this measurement campaign were levoglucosan and benzo(a)pyrene, or BaP, both being widely used as indicators for wood combustion in the field. (Saarnio et al., 2012)

The Aerosol Mass Spectrometer (AMS) onboard Sniffer was used to detect these markers during the measurements; additionally, the concentrations of these tracers were gotten from the HSY. As only graphs and not the actual data of the AMS or the HSY measurements were not available, most of the identification was done by analyzing the particle size distributions.

It has been previously shown that different particle sources produce characteristically different particle size distributions (Kristensson et al., 2013; Jamriska and Morawska, 2001). For example, wood combustion is known to produce particles with a diameter of around 0.1 μm. (Tissari et al., 2005) Diesel engines are commonly known to produce large numbers of very small particles, while street dust is dominantly very large particles (Vallero, 2008). In this manner the characteristic size distributions can be utilized for source attribution. An advanced form of source attribution with the same principal idea is to do a Positive Matrix Factorization, but this method is outside the scope of this thesis.
4.1 Weather

The amount of pollutants in the residential areas is dependent on the temperature as most wood combustion appliances, not including sauna stoves, are used for auxiliary heating in the Helsinki metropolitan area. (Myllynen, 2010) The coldest average daily temperature measured was -6.2 °C, a relatively mild temperature that does not cause considerable need for auxiliary heating. The temperature range during the measurement period was from -12 °C to +2 °C. Mostly the temperatures stayed between -6 to 0 °C. This temperature range was too mild to enable formation of inversion layer, and very effective in promoting slush formation and snowfall. The temperature graph covering the whole measurement period is presented in Appendix 3.

During the measurement period snowfall occurred on several occasions. Snowfall promotes particle deposition in the form of wet deposition. The snowfall estimates were only recorded verbally, as no applicable instrumentation was available. The snowfall was quite heavy at times during the measurement period. The snowfall was so severe on the 19th that measurements had to be cancelled on that day. Lighter snowfall occurred during most of the days in the measurement period and even this presumably had some effect on the measured concentrations.

4.2 Long-range transport of pollution

According to the definition of the OECD (2001), long-range transport is “the atmospheric transport of air pollutants within a moving air mass for a distance greater than 100 kilometers.” A typical LRT episode in the Helsinki Metropolitan region is one where pollutants come in from Eastern Europe (Malkki et al., 2012). This was the case at the beginning of the measurement period, when a LRT episode was present in the Metropolitan region from the 15th to the 20th. During this period the air masses moved in over the Baltic region and from Eastern Europe, as shown in Figure 7. The figure presents a trajectory from the 17th of February showing the path that the air mass traversed before arriving in southern Finland on that day.
The air mass was highly polluted and dominated the air quality in the entire metropolitan area as shown in Figure 8, which presents the PM$_{2.5}$ measurements by HSY. As can be seen in the figure, the PM$_{2.5}$ concentrations were clearly elevated in all measurement locations in the metropolitan region for the duration of the LRT episode. Small additional peaks can be found on the evening of 25$^{th}$ and 27$^{th}$ of February, suggesting some minor presence of LRT also on those days.
Figure 8 by HSY shows how the magnitude of the effect the LRT has on the air quality. The 24 hour average PM$_{2.5}$ limit of 25 µg/m$^3$ set by the World Health Organization was exceeded on the 15$^{th}$, 16th, 18th and 19$^{th}$. (HSY, 2013c) Many of the measurement stations of the HSY measured their highest yearly concentrations of various pollutants during the period between the 15$^{th}$ and the 20$^{th}$ of February. (Malkki et al., 2012)

The polluted air mass had a high concentration of PM$_{2.5}$ and PM$_{10}$. Also Levoglucosan and to some extent BaP were present, indicating an organic source and also further complicating the analysis of the days while LRT was present. (HSY, 2012) Due to the reasons stated above, the days with LRT present were discarded in the data analysis.

4.3 Particle Measurements

As mentioned above, information concerning the origin of particles can be gained by studying their size distribution. This can also be done by using sophisticated statistical tools like positive matrix factorization (PMF), which is in extensive use in the field (IE-HIAS, ca. 2010). Due to the limited extent of this thesis, only simpler methods were adopted here.

Figure 9, by Krecl (2008), shows the differing particle distributions that are produced by different sources. The figure, although being from a model used by Kreckl in PMF, depicts well the idea of source attribution by size distribution. Typical in situ measured
mode of RWC would fall around 80 – 160 nm, thus being somewhat larger than those used in the model by Krecl. (Tissari, 2005)

![Graph showing size distributions from Traffic, Residential Wood Combustion (RWC) and RWC and Long-Range Transport (LRT) as measured by Krecl (2008).]

Figure 9. The size distributions from Traffic, Residential Wood Combustion (RWC) and RWC and Long-Range Transport (LRT) as measured by Krecl (2008).

Traffic emissions have their maximum number concentration in the ultrafine region, as shown in Figure 9, sloping off quickly as the particle size grows. The RWC has only a limited amount of the ultrafine particles and forms a characteristic mode around 0.1 μm, while the LRT is mostly made up of larger, up to 1 μm particles.

Due to the limits of the measurement resolution of the Elpis, it was not possible to see the different modes as clearly as depicted in the model above. Nevertheless, the measurement of this thesis showed similar findings. The size distributions shown below in Figure 10 are from the actual measurements, and thus the different modes are merged together, and not separated as in the model above.
Figure 10. Median particle size distributions of different emission sources. RWC, Traffic(G) and Traffic(D) are based on \( n \) number of events averaged peak concentrations, while Urban background and LRT are the based on the ambient background on \( n \) number of days. RWC, \( n = 6 \); Traffic(G)(gasoline), \( n = 6 \); Traffic(D)(Diesel), \( n = 7 \); Urban Background, \( n = 5 \); LRT, \( n = 3 \).

The different emissions sources were identified based on the logbook records. The distributions shown in Figure 10 are based on observations from the logbook of traffic and presence of RWC. Two different size distributions were observed resulting from traffic, and the assumption was made that the different size distribution might be due to the different emission resulting from gasoline and diesel vehicles. This assumption is held throughout this report.

Despite the modes not being separated, it can still be clearly seen that the different emission sources had highly individual and recognizable size distributions. RWC was identified by having a distinct mode around 0.1 μm, and lacking in smaller particles. The gasoline cars were observed having a high particle concentration in the ultrafine region and a low concentration of larger particles. The diesel engines were identified by their mode that was between 30 and 90 nm and by the high particle concentrations in all size ranges.
LRT was also found to have very high concentration all across the range. The LRT also had the highest number of larger, around one micron, particles as would be expected due to the long residence time in the atmosphere and the growth resulting from that. The size distributions found had overall similar characteristics to those found in previous studies by Jamriska and Morawaska (2001) and Ellerman et al. (2009).

The comparison of the two Elpi’s gave information on the particle volatility as the other one was operated behind a thermal denuder at around 265 C° with the purpose of evaporating all volatile fractions of the particulate matter. Figure 11 below shows that most of the particles resulting from RWC were found to be nonvolatile, as the thermal treatment of the samples had no significant effect on the distribution. Also diesel emissions were not very strongly influenced by the TD. On average, the thermal treatment of RWC emissions resulted in only a 3 % decline in particle number, and a 14 % decline in particle volume concentration in the submicron particle region. The thermal treatment had a considerably stronger impact on traffic emissions than on the RWC.

![Particle Volatility](image)

Figure 11. Comparison of the two ELPIs median results over various events. High volatility can be found especially in the ultrafine particle region. The figure depicts the same observations as shown in figure 10.

Based on our measurements the TD reduced the particle number concentration for gasoline automobiles by 76 % and for diesel vehicles by 32 %. The volume concentrations of PM₁ in gasoline and diesel exhausts were reduced by 50 % and 44 %, respec-
tively. Previous studies have shown that most nucleation particles resulting from traffic are highly volatile, and the results of this thesis seemed to concur with this notion (Marjamäki and Keskinen, 2001).

The LRT also had a significant fraction of volatiles, and after the thermal treatment the total number-concentration decreased by 43 % and the volume concentration of PM$_1$ by 63 %. After thermal treatment, the size distribution of the LRT samples resembled that of RWC, suggesting a biomass based source, as can be seen in Figure 11. This notion is supported by the HSY measurements that showed the LRT having a significant concentration of wood combustion tracers also indicating a biomass based source (HSY, 2012).

Due to the thermal treatment, the submicron particle number and volume concentration of background air were reduced by 43 % and 75 %, respectively. This would suggest that a large fraction of the particles in the background are older particles which have had the time to grow in the atmosphere, as the ratio of volume to number loss is the largest here.

4.4 Gaseous pollutants

The biggest source for NO$_x$ in the metropolitan area is traffic according to Malkki et al. (2012). As expected, most of what was detected with the gas measurement devices were emissions from traffic. The gases that were measured during the routes were NO, NO$_2$, NO$_x$, CO and CO$_2$.

The city center and highways showed constantly significantly higher concentrations of NO$_x$'s and CO$_2$ than the residential areas. CO was the exception with fluctuating concentration. CO was observed having elevated concentrations on the western routes in comparison to the eastern ones, as can be seen in Appendix 5. The source of this elevation was unclear, as the NO and NO$_2$ levels were not elevated, ruling out traffic as the source. Most other measured pollutants exhibited an inverse trend, with higher concentrations found in the east rather than in the west.

Overall, RWC had only a minor effect on NO, NO$_2$ and CO$_2$ in comparison to regular automobile exhaust. This is demonstrated in Appendix 1, as a graph of measurements made on 26 February in Lintuvaara, Espoo. The observations shown in Appendix 1 are based on the measurement logbooks.
4.5 Urban background measurement

The baseline level of pollutants at the background measurement site was quite stable during the measurement period, excluding the LRT episode. In general, the days while the eastern routes were measured happened to coincide with days that had slightly elevated levels also in the urban background, making them seem more polluted. The measured background concentrations can be found in Appendix 5.

On the 25th, the background measurement levels were found to be elevated beyond typical variation. According to Malkki et al. (2012), the regional effects of RWC can be significant. This might explain the elevated levels of 25th, but it is more likely that there was also a component of LRT present, as the trajectories from that day show that the air masses had been moving into Finland over eastern Estonia and Russia.

4.6 Residential areas

The levels found at the urban background and the residential areas were quite similar, with the urban background site mostly having slightly lower concentrations overall. On weekends the pattern was stronger as there was less traffic influencing the background site and more wood combustion in the residential areas.

The areas were found to have rather similar air quality. Figure 12 shows a graph depicting the average particle size distributions of the six residential areas. Although the figure seems to indicate that the eastern routes have slightly higher concentrations, this is again due to the daily variation as the same trend was visible in the background measurements that were all made in Hietaniemi.
Figure 12. Median size distributions of the measured residential areas, along with the median background results for eastern and western routes separately.

The ultrafine region was found to be elevated in the eastern routes with respect to the background level. No such trend was found on the western routes, indicating possibly higher traffic rates in the eastern residential areas. The western routes showed a significantly higher concentration of particles around 80-300 nm which was not present in the background. This is likely to be due to wood combustion, as no other logical particle sources were present.

Table 2 gives the average concentration of chosen pollutants measured from the residential areas and separate background measurements for east bound and west bound days in order to take into account the daily variations of air quality. The background concentrations have not been removed from the table values, but are provided to enable comparison.
Table 2. Median measured concentrations of chosen pollutants within the residential areas and the urban background site for all of the measurement data collected within the areas.

<table>
<thead>
<tr>
<th>Location</th>
<th>NO</th>
<th>NO₂</th>
<th>PM₁</th>
<th>PM₂.₅</th>
<th>Ntot</th>
<th>CO</th>
<th>CO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ppb</td>
<td>ppb</td>
<td>µg/m³</td>
<td>µg/m³</td>
<td>1/cm³</td>
<td>ppm</td>
<td>ppm</td>
</tr>
<tr>
<td>Itä-Hakkila</td>
<td>0.9</td>
<td>8.4</td>
<td>17</td>
<td>15</td>
<td>11660</td>
<td>0.30</td>
<td>395</td>
</tr>
<tr>
<td>Päiväkumpu</td>
<td>0.7</td>
<td>7.1</td>
<td>22</td>
<td>19</td>
<td>8700</td>
<td>0.34</td>
<td>393</td>
</tr>
<tr>
<td>Vartiokylä</td>
<td>0.7</td>
<td>12.7</td>
<td>20</td>
<td>17</td>
<td>8130</td>
<td>0.13</td>
<td>397</td>
</tr>
<tr>
<td>Background (East bound days)</td>
<td>0.7</td>
<td>7.6</td>
<td>18</td>
<td>15</td>
<td>6520</td>
<td>0.05</td>
<td>391</td>
</tr>
<tr>
<td>Kattilalaakso</td>
<td>0.4</td>
<td>5.3</td>
<td>4</td>
<td>6</td>
<td>4950</td>
<td>0.58</td>
<td>391</td>
</tr>
<tr>
<td>Lintuvaara</td>
<td>0.9</td>
<td>4.4</td>
<td>4</td>
<td>5</td>
<td>5060</td>
<td>0.24</td>
<td>393</td>
</tr>
<tr>
<td>Laaksolahti</td>
<td>0.7</td>
<td>6.4</td>
<td>5</td>
<td>6</td>
<td>4740</td>
<td>0.60</td>
<td>393</td>
</tr>
<tr>
<td>Background (West bound days)</td>
<td>0.2</td>
<td>5.5</td>
<td>1</td>
<td>2</td>
<td>4840</td>
<td>0.05</td>
<td>395</td>
</tr>
</tbody>
</table>

The PM₁ and PM₂.₅ concentrations were slightly higher, along with CO in the western routes, possibly indicating wood combustion as the source. NO₂ and total particle number seemed to be higher in the eastern routes and would imply higher impact from traffic. Both observations also agree with the size distribution measured in the areas, as shown in Figure 12.

The mean values, with their standard deviations can be found in Appendix 4. According to these values, the nitrogen oxides seemed to be higher in the western route, and the PM₁ and PM₂.₅ had higher levels in the eastern route. However, the mean is quite far from the median, and with a very high standard deviation, indicating a possibly large error in the mean values.

4.6.1 Source attribution

In previous studies done in Denmark pollution source attribution has been done by developing a wood smoke profile. The wood smoke profile is a characteristic group of emissions that are produced by wood combustion, and it consists of particle number and volume, PM₂.₅ and soot parameters. This method has been used successfully as an indicator of wood combustion by Wåhlin et al. (2012)

Similar approach for the identification and classification of the different emission sources was used here. RWC was identified by a strong presence of PM₁ and PM₂.₅ and by looking at the particle size distribution, where wood combustion has its mode around 120 nm. Also the lack of the very high concentration of ultrafine particles was
required to rule out traffic. In addition, traffic was ruled out by looking at the levels of emitted NO\textsubscript{x}, and especially NO\textsubscript{x}, which would rather indicate internal combustion engine as the source if high concentrations were found.

The study by Wåhlin et al. (2010) also used the ratio of between PM\textsubscript{2.5} and NO\textsubscript{x} and total submicron particle number (N\textsubscript{tot}) and NO\textsubscript{x} to quantify the proportion of particles resulting from traffic. They found that traffic produced PM\textsubscript{2.5}/NO\textsubscript{x} ratio of 0.08 μg/ppb and N\textsubscript{tot}/NO\textsubscript{x} ratio of about 300 1/ppb.

The measurements performed for this study found quite similar results. From the measurements in the city center, where most submicron particles result from vehicle exhaust, this thesis determined ratios of 0.11 μg/ppb for PM\textsubscript{2.5}/NO\textsubscript{x} and 305 1/ppb for N\textsubscript{tot}/NO\textsubscript{x} for traffic. Figure 13 depicts the ratios of PM\textsubscript{2.5} and PM\textsubscript{1} to NO\textsubscript{x} measured during the campaign for each measurement area.

![Figure 13: Ratios of PM\textsubscript{2.5}/NO\textsubscript{x} and PM\textsubscript{1}/NO\textsubscript{x} measured during the campaign at different locations.](image)

It can be seen that the residential areas all had significantly higher PM to NO\textsubscript{x} ratios than did the city or the highways. Also the PM\textsubscript{1}/NO\textsubscript{x} ratio was higher in the residential areas than even in the background, suggesting an external source besides traffic for the particles. Although traffic produced large numbers of particles, it also produced a lot of NO\textsubscript{x}\textsubscript{s}, as shown by the low PM\textsubscript{2.5} to NO\textsubscript{x} ratios. The high PM\textsubscript{2.5} to NO\textsubscript{x} ratios measured at the residential areas indicates additional particle sources that do not produce high amounts of NO\textsubscript{x}\textsubscript{s}. RWC could be a viable source for these particles.
The campaign measurements show clearly that RWC deteriorates air quality significantly. It produces large concentrations of PM$_1$ and PM$_{2.5}$, and elevates the CO levels. It also causes a slight elevation in NO and NO$_2$, but the effect is minor in comparison to that caused by vehicles. While it was not feasible in the extent of this thesis to numerically determine the proportion of pollutant resulting from RWC, the numbers that are reported by Saarnio et al. (2012), 18-60 % seem realistic.

4.6.2 Temporal distribution

Air pollution and air quality are highly dependent on time. The morning and afternoon rush-hours are a clear everyday example of this. Wood combustion also occurs in this bimodal pattern, with one peak in the morning and a larger one in the evening. (Myllynen, 2010) The morning’s peak is usually considerably smaller and occurs before people leave for work. Due to the smaller impact of the morning peaks, the measurements focused on the evening peaks. The averaged concentrations for morning and evening measurements are given in the Appendix 2.

Pollution concentrations were observed to be significantly smaller in the morning than in the evening. As the evening progressed, the NO emissions were observed to decrease gradually, probably due to the decreased traffic. The same trend appeared with NO$_2$, but to a smaller extent, as can be seen in the Appendix 2. Particle emissions were observed increasing towards the evening, with a noticeable difference in the range of > 100 nm, coinciding with the typical wood combustion mode. The same increasing trend was apparent in both PM$_1$ and PM$_{2.5}$ concentrations that showed growth toward the latter part of the evening, being highly suggestive of wood combustion as traffic levels had already tended to decline at this point.

The wood combustion rate also varies on a week/weekend cycle, where usually the concentration is much higher during the weekends. According to a previously done poll on wood combustion, people burn wood more often during the weekend than during the week. Especially Wednesdays and Saturdays have more wood combustion, due to these being traditional days for heating the sauna in Finland. (Myllynen, 2010)

Due to the disruption caused by the LRT episode, it was possible to measure reliably only during one weekend. Also one Wednesday was measured, but it was heavily influenced by the LRT. The measurement results were mostly in accordance with the
poll, and higher concentrations were found in the east during the weekend than during the week days. The found particle size distributions are shown in Figure 14. It should be noted that here the weekend includes the evening but not the morning of Friday the 24th.

Figure 14. Different median particle size distributions of the residential areas during the week and weekends in the Eastern and Western routes recorded during the measurement period.

As can be seen above, the concentration of the ultrafine particles in the western route was quite similar during the week and weekends. There is however some change in the larger particles that would fit the mode of wood combustion. The trend is harder to read from the eastern route, as the difference is so large across all size ranges. The eastern weekend is based only on measurements made on the 25th, and as mentioned before some LRT was present during that day. This makes it extremely hard to estimate how much is due to the LRT and how much is caused by the increased wood combustion of Saturday evening.

4.6.3 Spatial distribution within the residential areas

The spatial distribution of pollutants is dependent on multiple factors. Previous studies have shown that particles resulting from RWC can either, stay in a localized area, be
transported regionally or even become long range pollution (Saarnio et al., 2012). According to the HSY, the spatial variation of pollutants might be highly significant even with in a residential area, and the results can be strongly dependent on the measurement location (Malkki et al., 2012).

The Sniffer vehicle has an excellent ability to catch the spatial distribution of the pollutants. It is often difficult to tell from stationary measurement locations the applicability of the data to a larger area. It might be that there is a single house next to the measurement station emitting high levels of pollutants that are then thought to represent the entire measurement area. Figure 15 depicts the map of Kattilalaakso, one of the monitored residential areas in the westbound route, where HSY had a stationary air quality measurement site. The arrow shows the locations of the site.
As can be seen in Figure 15, the air quality can vary very sharply as a function of distance. Multiple strong peaks that might have an order of magnitude difference in concentration can fit in the same stretch of road. Although most of these sharp peaks are due to traffic, especially snow ploughs or busses, also wood combustion was found to have very dramatic effects, however, in limited areas.

Wood combustion has especially large impact on the volume concentration of particles, as it produces larger particles than traffic. Single smokestack can momentarily change the order of magnitude for the total particle volume concentration of submicron particles. Figure 16 depicts the particle volume distribution calculated from the Elpi data for...
car and wood combustion emissions as well as the distribution of the LRT and typical background in the residential areas.

![Graph showing median volume distributions of RWC, Traffic, Traffic (D), RB, and LRT](image)

**Figure 16.** Median volume distributions of RWC (detected wood combustion events) \((n=6)\), Traffic (Gasoline vehicles) \((n=6)\), Traffic (Diesel vehicles) \((n=6)\), RB (overall residential areas) and LRT \((n=5)\). Calculated using spherical assumption for the particle shape.

It can be seen in Figure 16 how the relatively few larger particles dominate the volume concentration, and thus often also mass concentration. The extremely high numbers of < 50 nm particles that are emitted from cars have practically no effect on the volume or mass concentrations.

With the mobile measurement capabilities of the Sniffer vehicle, one can identify local variations and measure prevailing average conditions of an area. This can be done with much higher confidence than is possible from fixed measurement sites, which can be vulnerable to local variations that, as shown above, can be high.

The data produced by the fixed measurement sites, mentioned above, was compared to the data collected during the measurement loops of Sniffer. The data that was avail-
able from the HSY’s measurement stations was limited to NO, NO₂ and PM₂.₅ in both Vartiokylä and Kattilaakso. As shown in Figure 17, the data showed a strong positive correlation between the stationary measurement sites of HSY and the mobile measurements performed with Sniffer. Especially the NOₓ and PM₂.₅ were found to have a strong correlation. The slope should be noted as the mobile measurements showed consistently higher concentrations than did the stationary measurements.

The correlation between the NO measurements is not as good as with PM₂.₅, NO₂ and NOₓ. The correlation with NO was the only one without a statistically significant or highly significant p-value. This variability is most probably explained by the rapid oxidation process of NO in atmospheric conditions. Sniffer measured significantly higher levels of NO than the HSY’s stationary measurement sites did, which would be expected as it measures direct emission from passing cars. By the time the stationary measurement post record the emitted NO, most of it has already converted to NO₂. This is supported by the fact that NOₓ showed stronger correlation than did NO or NO₂ by themselves.
Overall, a very strong correlation was found between the stationary measurement data, and that recorded by Sniffer. Despite the potential vulnerability to distortions by emissions sources in the immediate vicinity of the measurement site, the fixed measurement stations seemed to produce a highly representative picture of the general air quality in a given area.

5 Conclusions

The pollutant with the largest impact on human health in the Helsinki metropolitan region is currently fine particles. In recent years, the sources and effects of fine particles have been given an increasing amount of attention and their effects have been somewhat better understood. One of the major sources for fine particles that have been identified in these studies is residential wood combustion. Estimates on the proportion of particles produced by RWC in the Helsinki region vary around 15-60 per cent. (Malkki et al., 2012; Saarnio et al., 2012)

In the Helsinki region wood is mostly used as auxiliary heating, apart from the sauna stoves that see regular use throughout the year (Myllynen, 2010). Due to the mild temperature during the measurement period, there was only little need for auxiliary heat sources, which diminished the need for wood combustion and thus lowering the emission levels present. Also because of the mild weather there was no chance for an inversion layer to form, which would have trapped the particles in the area.

First part of the measurement period coincided with a LRT episode, which when present, was found to be the dominant factor for most pollutants. Mostly the ultrafine particles and NO, which both have short lifespan in the atmosphere, were impacted more by the local actions.

The measurements performed with the Sniffer vehicle show very high local variations within the residential areas. Most of the variation is due to either traffic emissions, such as cars passing by, or emissions from a single chimney. When the data is averaged over hours, the concentrations measured by Sniffer seem to agree to a high extent with those that the HSY measures. Apparently the long sampling period used by the HSY naturally averages out these short term variations within the micro environments.
The measurements also show that a single chimney can significantly decrease the air quality for a highly localized area, with the effect lasting up to multiple hours as wood combustion usually is a long event. The effects of these local point sources were clearly visible as repeating patterns of high concentrations, on occasions when routes were driven more than once in an evening.

The six residential areas that were monitored were all found to have quite similar air quality amongst each other and to that of the urban background. Due to the limited length of the measurement period, it was impossible to determine if some micro-environments existed within the residential areas. The noise in the data produced by the cars, in addition to the limited number of measurement days, made these kinds of subtle differences indistinguishable from the background variations.

Although the estimation of exact effects of RWC on air quality is outside the extent of this thesis, it is clear from our measurements that RWC has a highly significant impact on the overall air quality. This can be seen in the measured particle size distributions, which had a clear presence of particles ranging from 100-200 nm, which coincides with the mode resulting from wood combustion.

On the basis of the measurements, it seems that RWC has larger impact on the particles, rather than gaseous pollutant, although elevated levels of CO were usually found coinciding with RWC. Even during the peak hours of RWC, Saturday evening, the main source for NOx’s and other gaseous pollutants in the suburban areas remained traffic.

The findings show that RWC produces a recognizable characteristic emission with an elevated level of particles with a diameter of around 120 nm present, accompanied by high PM1 and PM2.5 concentrations, and a relatively low amount of NO. The particles produced by RWC also showed low volatility, with a 3 % decrease in particle number. In comparison, traffic emissions produced an extremely high particle count with a mode around 10-40 nm, and showed high volatility. A decline in particle number up to 76 % for gasoline and 32 % for diesel vehicles was measured. Traffic emissions were found to be accompanied also by high NOx emissions and relatively low concentrations of PM1 and PM2.5.

The largest problem remains the limited amount of measurement days that was further limited by the presence of LRT during the first days. The limited length with suboptimal weather and road conditions created unavoidably some uncertainty into the data, but
the very strong correlation with the HSYs stationary measurement results even in the harsh winter conditions did not affect the validity of the data produced.
References


Environnement S.A., 2013. CO12M. [online] Available at: <http://www.environnementsa.in/products/co12m>


Helsinki Regional Environmental Services, 2011. MMEA PKS-mittausten suunnittelu 2. kausi. [meeting memo] November 2001. HSY.


Helsinki Regional Environmental Services, 2013c. Pienhiukkaset viikoittain. [online] Available at:


Appendices

Appendix 1: Example of RWC and Traffic Emissions

26.2.2012, Lintuvaara, Espoo

![Graph showing pollution levels over time]

- NO
- NO2
- NOx
- CO2

Cars
Wood Combustion

Time: 19:52:20 to 20:12:20
Levels: -10 to 150 ppb
Appendix 2: Effects of temporal variation on air quality

Average Particle Size Distributions of the Residential Areas

PM and NOx concentrations in the residential areas from the 20th to the 27th

NO

NOx

NO2

PM 2.5

PM 1
Appendix 3: Temperature during the measurements

Lämpötila 15.02.–27.02.2012

°C

16.02 17.02 18.02 19.02 20.02 21.02 22.02 23.02 24.02 25.02 26.02 27.02

Pasila Åmlassuo Liukki
### Appendix 4: Mean pollutant concentrations measured in the residential areas

<table>
<thead>
<tr>
<th>Location</th>
<th>NO ppb</th>
<th>NO₂ ppb</th>
<th>PM₁ ug/m³</th>
<th>PM₂.₅ ug/m³</th>
<th>Ntot 1/cm³</th>
<th>CO ppm</th>
<th>CO₂ ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Itä-Hakkila</td>
<td>10.0</td>
<td>14.2</td>
<td>22.5</td>
<td>20.0</td>
<td>14261</td>
<td>0.4</td>
<td>394.6</td>
</tr>
<tr>
<td>Itä-Hakkila SD</td>
<td>37.2</td>
<td>39.1</td>
<td>24.5</td>
<td>18.5</td>
<td>25091</td>
<td>0.3</td>
<td>6.8</td>
</tr>
<tr>
<td>Päiväkumpu</td>
<td>4.4</td>
<td>9.0</td>
<td>25.6</td>
<td>22.8</td>
<td>11447</td>
<td>0.4</td>
<td>393.2</td>
</tr>
<tr>
<td>Päiväkumpu SD</td>
<td>13.2</td>
<td>9.4</td>
<td>29.0</td>
<td>22.1</td>
<td>10383</td>
<td>0.2</td>
<td>6.3</td>
</tr>
<tr>
<td>Vartiokylä</td>
<td>6.5</td>
<td>15.8</td>
<td>21.7</td>
<td>18.8</td>
<td>10500</td>
<td>0.1</td>
<td>397.0</td>
</tr>
<tr>
<td>Vartiokylä SD</td>
<td>20.2</td>
<td>22.3</td>
<td>20.6</td>
<td>15.4</td>
<td>8654</td>
<td>0.3</td>
<td>10.3</td>
</tr>
<tr>
<td>Background (East bound days)</td>
<td>2.7</td>
<td>12.3</td>
<td>13.5</td>
<td>13.2</td>
<td>6726</td>
<td>0.0</td>
<td>395.1</td>
</tr>
<tr>
<td>Background SD (East bound days)</td>
<td>6.9</td>
<td>12.7</td>
<td>9.0</td>
<td>5.6</td>
<td>2739</td>
<td>0.1</td>
<td>7.1</td>
</tr>
<tr>
<td>Lintuvaara</td>
<td>4.3</td>
<td>6.3</td>
<td>10.0</td>
<td>10.0</td>
<td>6167</td>
<td>0.5</td>
<td>390.4</td>
</tr>
<tr>
<td>Lintuvaara SD</td>
<td>18.9</td>
<td>10.2</td>
<td>41.9</td>
<td>31.6</td>
<td>9723</td>
<td>0.2</td>
<td>3.7</td>
</tr>
<tr>
<td>Kattilalaakso</td>
<td>17.7</td>
<td>13.3</td>
<td>6.3</td>
<td>6.7</td>
<td>14259</td>
<td>0.3</td>
<td>396.0</td>
</tr>
<tr>
<td>Kattilalaakso SD</td>
<td>48.2</td>
<td>30.1</td>
<td>11.7</td>
<td>9.8</td>
<td>28894</td>
<td>0.2</td>
<td>9.8</td>
</tr>
<tr>
<td>Laaksolahti</td>
<td>6.5</td>
<td>10.4</td>
<td>9.3</td>
<td>9.2</td>
<td>5879</td>
<td>0.6</td>
<td>392.7</td>
</tr>
<tr>
<td>Laaksolahti SD</td>
<td>26.1</td>
<td>21.7</td>
<td>19.0</td>
<td>14.6</td>
<td>5103</td>
<td>0.4</td>
<td>6.0</td>
</tr>
<tr>
<td>Background (West bound days)</td>
<td>-1.9</td>
<td>5.0</td>
<td>2.7</td>
<td>4.2</td>
<td>4489</td>
<td>0.1</td>
<td>393.7</td>
</tr>
<tr>
<td>Background SD (West bound days)</td>
<td>12.7</td>
<td>8.7</td>
<td>3.4</td>
<td>3.7</td>
<td>2006</td>
<td>0.0</td>
<td>5.3</td>
</tr>
</tbody>
</table>
Appendix 5: Measurement results by area

1st column of 21st and 24th are morning measurements from 9 to 12, while all other evening measurements from 17 to 23.

[Graph and table as described in the text]
### Appendix

#### Table: Urban Air Quality Indices

<table>
<thead>
<tr>
<th>Date</th>
<th>Avg. Temp (°C)</th>
<th>RH (%)</th>
<th>City</th>
<th>Nloc (1/cm³)</th>
<th>PM 1 (mg/m³)</th>
<th>PM 2.5 (mg/m³)</th>
<th>NO2 (ppb)</th>
<th>NO (ppb)</th>
<th>CO (ppm)</th>
<th>CO2 (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>15.2</td>
<td>75.7</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>16.2</td>
<td>71.6</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>17.2</td>
<td>74.3</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>18.2</td>
<td>68.6</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>19.2</td>
<td>71.4</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>20.2</td>
<td>84.3</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>21.2</td>
<td>76.2</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>22.2</td>
<td>73</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>23.2</td>
<td>75.3</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>24.2</td>
<td>75.8</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
<tr>
<td></td>
<td>25.2</td>
<td>69.5</td>
<td></td>
<td>1.0</td>
<td>0.1</td>
<td>0.001</td>
<td>-100</td>
<td>60</td>
<td>200</td>
<td>300</td>
</tr>
</tbody>
</table>
### Itä-Hakkila = White, Laaksolahti = Grey

<table>
<thead>
<tr>
<th>Date</th>
<th>Avg. Temp.</th>
<th>RH</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.2</td>
<td>-39</td>
<td>37.5</td>
</tr>
<tr>
<td>16.2</td>
<td>-2</td>
<td>87.6</td>
</tr>
<tr>
<td>17.2</td>
<td>-4.2</td>
<td>81.9</td>
</tr>
<tr>
<td>18.2</td>
<td>-4.1</td>
<td>16.1</td>
</tr>
<tr>
<td>19.2</td>
<td>-3</td>
<td>80.8</td>
</tr>
<tr>
<td>20.2</td>
<td>1.8</td>
<td>82</td>
</tr>
<tr>
<td>21.2</td>
<td>-1.9</td>
<td>78.9</td>
</tr>
<tr>
<td>22.2</td>
<td>-1.8</td>
<td>81.7</td>
</tr>
<tr>
<td>23.2</td>
<td>-2.7</td>
<td>86.7</td>
</tr>
<tr>
<td>24.2</td>
<td>-2.6</td>
<td>71.9</td>
</tr>
</tbody>
</table>

### Concentrations

<table>
<thead>
<tr>
<th>Date</th>
<th>N\text{tot} (1/cm³)</th>
<th>PM 1 (mg/m³)</th>
<th>PM 2.5 (mg/m³)</th>
<th>NO2 (ppb)</th>
<th>NO (ppb)</th>
<th>CO (ppm)</th>
<th>C02 (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>15.2</td>
<td>1e+05</td>
<td>0.1</td>
<td>0.1</td>
<td>20</td>
<td>10</td>
<td>2</td>
<td>450</td>
</tr>
<tr>
<td>16.2</td>
<td>1e+04</td>
<td>0.1</td>
<td>0.1</td>
<td>10</td>
<td>5</td>
<td>1</td>
<td>420</td>
</tr>
<tr>
<td>17.2</td>
<td>1e+03</td>
<td>0.01</td>
<td>0.01</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>390</td>
</tr>
</tbody>
</table>