Air pollution and health –
Indicators, trends and impacts

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Academic dissertation for the Degree of Doctor of Philosophy in Applied Environmental Science at Stockholm University to be publicly defended on Thursday 13 June 2019 at 10.00 in Nordenskiöldsalen, Geovetenskapens hus, Svante Arrhenius väg 12.

Abstract
This thesis focuses on some of the limitations and difficulties that exist when it comes to quantifying the human health effects that arise as a result of air pollution exposure. The following four issues are particularly analysed and discussed: 1) The measurement techniques used for carbonaceous particles and their lack of consistency; 2) Do the health risks associated with exposure to PM$_{10}$ depend on the content of elemental carbon in the aerosol?; 3) Trends in air pollutants and the health effects that arise as a result of changed exposure to the measured pollutants; 4) The associations between the measured concentrations of different air pollutants in Stockholm and the daily number deaths; 5) Air quality indicators and health outcomes as basis for an air quality health index (AQHI).

The method that has been used is largely based on empirical data analysis, where further statistical processing has been used in order to clarify the scientific issues. The overall conclusions are the following: 1) The health impact assessments associated with exposure to carbonaceous particles would benefit from the introduction of a more uniform measurement technique in order to get more consistent and reliable results; 2) The health risks associated with exposure to PM$_{10}$ are dependent on the content of elemental carbon; 3) The life expectancy increase associated with decreasing NO$_x$ trends during 1990–2015 in Stockholm, Gothenburg and Malmo make up as much as about 20 % of the total gain in life expectancy during this period, which clearly shows the beneficial effects related to decreased exposure; 4) The associations between daily mortality and the concentrations of O$_3$ and PM$_{2.5-10}$ in Stockholm are statistically significant, which does not apply to the exhaust-related pollutants, possibly reflecting behavioural factors affecting the degree of exposure; 5) In comparison with the currently used air quality index (AQI), the air quality health index (AQHI) is a more useful tool in order to address the short-term health effects associated with multi-pollutant exposure to NO$_x$, O$_3$, PM$_{10}$ and birch pollen.

We hope that these findings will be useful from a policy point of view. Introducing a more consistent measurement technique for soot particles would be beneficial in assessing the health effects related to exposure to these particles. The increase in life expectancy associated with decreasing NO$_x$ trends shows the benefits from a public health perspective when it comes to introducing emission-reducing measures from traffic. The AQHI would be beneficial to implement in legislation, as it is based on several pollutants, which means that the cumulative health effects associated exposure to several different air pollutants are accounted for.

Keywords: air pollution, health, measurements, trends, impacts, mortality, NO$_x$, elemental carbon, AQHI.
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Sammanfattning

Denna avhandling fokuserar på de begränsningar och svårigheter som finns när det gäller att kvantifiera de hälsoeffekter som uppstår till följd av luftföroringsexponering. Följande fyra frågor har i särskilt hög grad analyserats och diskuterats: 1) Mätteknikerna som används för att mäta sotpartiklar och deras brist på överensstämmelse; 2) Är de hälsoeffekter som är kopplade till exponering av PM$_{10}$ beroende av halten av elementärt kol (EC)?; 3) Trenden i luftföroringenshalter och de hälsoeffekter som uppstår till följd av förändrad exponering av de uppmätta föroreningarna; 4) Kopplingen mellan de uppmätta koncentrationerna av olika luftföroringar i Stockholm och det dagliga antalet dödsfall; 5) Luftkvalitetsindikatorer och hälsoutfall som utgångspunkt för ett hälsoindex för luftkvaliteten (AQHI).

Metoden som har använts bygger till stor del på analys av empirisk data, där fortsatt statistisk bearbetning har gjorts för att reda ut de vetenskapliga frågeställningarna. De övergripande slutsatserna är följande: 1) Hälsokonsekvensberäkningar gällande exponering för sotpartiklar skulle gynnas av införandet av en mer enhetlig mätteknik för att få mer konsekventa och tillförlitliga resultat; 2) Hälsoeffekterna som är kopplade till exponering av PM$_{10}$ är beroende av halten av elementärt kol; 3) Den ökning i genomsnittlig livslängd som är kopplad till sjunkande trender för NO$_x$ under perioden 1990–2015 i Stockholm, Göteborg och Malmö utgör upp till 20 % av den totala ökningen i förväntad livslängd under denna period, vilket tydligt visar de gynnsamma effekter som uppstår till följd av minskad exponering; 4) Sambanden mellan daglig mortalitet och halterna av O$_3$ och PM$_{2.5-10}$ i Stockholm är statistiskt signifikanta, något som inte gäller för de avgasrelaterade föroreningarna, vilket möjligt har att göra med beteendemässiga faktorer som påverkar graden av exponering; 5) I jämförelse med det luftkvalitetsindex som för närvarande används (AQI), är AQHI ett mer användbart verktyg för att hantera korttidseffekterna av samtidig exponeringen för flera föroreningar, där NO$_x$, O$_3$, PM$_{10}$ och björkpollen ingår.

Vi hoppas att dessa ovan nämnda resultat kan vara användbara från en politisk synvinkel. Att införa en mer konsekvent mätteknik för sotpartiklar skulle vara till fördel när det gäller att bedöma hälsoeffekterna till följd av exponering av dessa partiklar. En ökning av den förväntade livslängden i samband med sjunkande koncentrationer av NO$_x$ visar fördelarna ur ett folkhälsovPerspektiv när det gäller att införa utsläppsminskande åtgärder från trafiken. AQHI skulle vara fördelaktigt att implementera i lagstiftningen, eftersom detta index är baserat på flera föroreningar, vilket innebär att de kumulativa hälsoeffekterna kopplade till exponering av flera olika luftföroringar illustreras.
List of papers and authors contribution

I  The use of carbonaceous particle exposure metrics in health impact calculations
   
   H. Olstrup, C. Johansson, and B. Forsberg

   Published in International Journal of Environmental Research and Public Health (IJERPH)
   2016, 13(3), 249; doi:10.3390/ijerph13030249

II Trends in air pollutants and health impacts in three Swedish cities over the past three
decades
   

   Published in Atmospheric Chemistry and Physics, 18, 15705-15723, 2018
   https://doi.org/10.5194/acp-18-15705-2018

III Association between mortality and short-term exposure to particles, ozone and nitrogen
dioxide in Stockholm, Sweden
   
   H. Olstrup, C. Johansson, B. Forsberg, and C. Åström

   Published in International Journal of Environmental Research and Public Health (IJERPH)
   2019, 16(6), 1028; doi:10.3390/ijerph16061028

IV A multi-pollutant air quality health index (AQHI) based on short-term respiratory effects
in Stockholm, Sweden
   
   H. Olstrup, C. Johansson, B. Forsberg, A. Ekebom, K. Meister and A. Tornevi

   Published in International Journal of Environmental Research and Public Health (IJERPH)
   2019, 16(1), 105; doi:10.3390/ijerph16010105
Contribution of the author

Paper I
The author conducted a literature review and a synthesis of the previous research in this field. The author also wrote the manuscript and conducted, together with the co-authors, the calculations and the discussion of the results.

Paper II
The author wrote the manuscript and conducted in cooperation with the co-authors the calculations in Openair and AirQ+. Interpretation and discussion of the results were conducted in cooperation with the co-authors.

Paper III
The author wrote the manuscript and interpreted the results in cooperation with the co-authors.

Paper IV
The author wrote the manuscript and conducted the data analysis and the health impact calculations. Interpretation and discussion of the results was conducted in cooperation with the co-authors.
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Abbreviations

Abs  Absorbance
AEDV  Asthma emergency department visits
AQG  Air Quality Guideline
AQHI  Air quality health index
AQI  Air quality index
ARI  Aggregate risk index
\(\beta\)  Beta-coefficient
BC  Black carbon
BS  Black smoke
CI  Confidence interval
CPC  Condensation particle counter
D.F.  Degrees of freedom
EC  Elemental carbon
EUCAARI  European Integrated project on Aerosol, Cloud, Climate, and Air Quality Interactions
GBD  Global Burden of Disease
IQR  Interquartile range
K  Kelvin
LLE  Loss of life expectancy
Nm  Nanometre
NO\(_x\)  Generic term for nitrogen monoxide and nitrogen dioxide
NO  Nitrogen monoxide
NO\(_2\)  Nitrogen dioxide
O\(_3\)  Ozone
OC  Organic carbon
PAH  Polycyclic aromatic hydrocarbons
PC  Pyrolytic carbon
PM  Particulate matter
PM\(_{2.5}\)  Particles with an aerodynamic diameter smaller than 2.5 µm
<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
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<tbody>
<tr>
<td>PM$_{2.5-10}$</td>
<td>Particles with an aerodynamic diameter in the range of 2.5–10 µm</td>
</tr>
<tr>
<td>PM$_{10}$</td>
<td>Particles with an aerodynamic diameter smaller than 10 µm</td>
</tr>
<tr>
<td>PNC</td>
<td>Particle number count</td>
</tr>
<tr>
<td>PNC$_4$</td>
<td>Particles with an aerodynamic diameter larger than 4 nm</td>
</tr>
<tr>
<td>PNC$_7$</td>
<td>Particles with an aerodynamic diameter larger than 7 nm</td>
</tr>
<tr>
<td>RMSE</td>
<td>Root mean square error</td>
</tr>
<tr>
<td>R</td>
<td>Correlation coefficient</td>
</tr>
<tr>
<td>R$^2$</td>
<td>Coefficient of determination</td>
</tr>
<tr>
<td>RR</td>
<td>Relative risk</td>
</tr>
<tr>
<td>SC</td>
<td>Soot carbon</td>
</tr>
<tr>
<td>TEOM</td>
<td>Tapered element oscillating microbalance</td>
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<tr>
<td>UFP</td>
<td>Ultrafine particles</td>
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</table>
1. Introduction

The air that we inhale contains a large number of gases and particles, originating from both natural and anthropogenic sources. The health effects associated with exposure to these gases and particles vary throughout the spectrum of their composition; oxygen is vital for a large number of living organisms, while other gases, like carbon monoxide, are potentially dangerous to inhale. Anthropogenic particles are in most cases, to a greater or lesser extent, harmful to the human health. For many pollutants, there are large uncertainties regarding the causal links between exposure and different health outcomes. The fact that the air contains a large number of different air pollutants makes it more complex when it comes to evaluate individual effect from each pollutant. In order to quantify the health effects associated with air pollution exposure, there are a number of uncertainties, where there is a need for further investigations and analyses.

Since air pollution exposure poses a serious global health problem, it is important to increase the knowledge gaps in order to mitigate the health risks associated with air pollution exposure. According to the Global Burden of Disease (GBD) in 2016, PM$_{2.5}$ is estimated to be the sixth largest risk factor for premature deaths on the global scale (see Figure 1). Worldwide, exposure to PM$_{2.5}$ in the ambient air contributed to 4.1 million premature deaths from heart disease and stroke, chronic lung disease, lung cancer and respiratory infections. Ozone is also an important component of air pollution, and exposure to ozone contributed to 243 000 premature deaths worldwide during 2016 caused by chronic lung disease (Health Effects Institute, 2018). In Figure 1, a number of factors contributing to premature deaths are ranked. The diseases that are caused by the different factors are also specified.
Despite the fact that air pollution exposure constitutes a serious global health risk, there are still a lot of uncertainties that need to be addressed. This thesis is focused on some of the limitations and difficulties that there are when it comes to quantifying the health effects that arise as a result of air pollution exposure. The following five issues are particularly analysed and discussed: 1) The measurement techniques used for carbonaceous particles and their lack of consistency; 2) Are the health risks associated with exposure to PM$_{10}$ dependent on the amount of elemental carbon in PM$_{10}$?; 3) Trends in air pollutants and the health effects that arise as a result of changed exposure to the measured pollutants; 4) The associations between the measured concentrations of different air pollutants in Stockholm and the daily number of deaths; 5) Air quality indicators and health outcomes as basis for an air quality health index (AQHI).
2. Background and scientific goals of the thesis

When it comes to health effects associated with exposure to airborne particles, differences among the measurement techniques used is an important issue to address. Among all different kinds of air pollutants, particles are considered to be the factor that causes the greatest health impact in Europe (EEA, 2014). The legislation in Europe regulates undefined particulate matter (PM$_{2.5}$ and PM$_{10}$), despite that the results from many studies indicate that combustion-related carbon-containing particles are particularly important in terms of health effects related to exposure (Grahame et al., 2014; Janssen et al., 2011). From a health perspective, it would therefore be highly relevant to involve soot particles in the legislation. However, there are a lot of uncertainties which makes this change difficult to execute. The different kinds of measurement techniques, which all are intended to measure soot particles, have varied historically, and measurement methods also differ between countries and continents. When different cities and places are compared, the relationships between different soot measurement methods are not consistent. This means that there are difficulties in the recalculation of the relationship from one exposure measurement to another. The aim of the Paper I was to address the above mentioned issues, and to illustrate the uncertainties in health impact calculations associated with the recalculation from one measurement technique into another.

The aim of Paper II has been to quantify the health effects related to trends in air pollution concentrations in the largest cities in Sweden. There are two main reasons to do this; the first one is to get clarity on how air pollution levels have changed in recent decades, and the second one is to calculate the health benefits or detriments associated with changes in exposure. The change in life expectancy has been used as an indicator of the health impact associated with change in exposure. To get an indication of how important air pollution exposure is to public health, the changes in life expectancy associated with changes in exposure have been compared with the total gain in life expectancy that has occurred during the surveyed period of time. Another issue that has been addressed is the limitation of assessing the population exposure based on the measured levels at a centrally located measuring station. This is assessed by comparing model-calculated annual population-weighted exposure concentrations with annual-mean urban-background concentrations.

The aim of Paper III has been to analyse the effects on daily mortality associated with different air pollution indicators. Besides the usual pollutants, i.e. PM$_{10}$, PM$_{2.5}$, NO$_2$ and O$_3$, the analyses also include PNC$_4$, PM$_{2.5-10}$ and BC, which are not so common in epidemiological studies. The goal of this paper was to compare the effects on daily mortality associated with exposure to the different air pollutants, and also to explain the reasons for the results.

The aim of Paper IV was to address the combination effects resulting from simultaneous exposure to several different air pollutants. An air quality health index (AQHI), based on the short-term health effects associated with three air pollutants and birch pollen has been calculated for Stockholm, with the intention to illustrate the combination effect, and its annual variation.
3. Materials and methods

3.1. The measuring stations and their locations

The basic method that has been used is largely based on analysis of empirical data. In Paper I, data reported in different scientific publications have been used for further analysis. In Paper II, III and IV, data obtained from regulatory monitoring stations in three Swedish cities, representing urban-background concentrations, have been used for both the trend analyses and the health impact calculations. Statistical analyses have been performed in Excel, Openair, R and AirQ+.

The analyses in Paper II, III and IV are based on the results from measuring stations in Stockholm, Gothenburg and Malmö. These stations are centrally located in each city and represent urban background concentrations of the measured pollutants. The measuring station in Stockholm is located at Torkel Knutssonsgatan on a roof 20 m above ground level, in Gothenburg it is located on a roof 30 m above ground level in the neighbourhood Östra Nordstan, and in Malmö, it is located on a roof 20 m above ground level at the city hall (Rådhuset) in the city centre (Figure 2). They are all regulatory monitoring urban-background stations using reference methods for the measurements of NOx, NO2 and O3. The method that has been used for measuring PM₁₀ is equivalent to the reference method. For BC and PNC, there are no reference methods. Birch pollen have been measured on the roof of the Arrhenius Laboratories at Stockholm University.

![Figure 2. Stockholm, Gothenburg, and Malmo; the three largest cities in Sweden, and their surroundings. The black dot in each city shows the location of the measuring station. Each map represents an area of 35 x 35 km.](image)

3.2. Description of the measurement techniques

The measurement stations in Stockholm, Gothenburg and Malmö have all provided us with data regarding NOx, NO2, O3 and PM₁₀. In Paper III, data for PM₂.₅, PM₂.₅-₁₀, BC and ultrafine particles (PNC₄ and PNC₇) have also been obtained from the measuring station at Torkel Knutssonsgatan in Stockholm. In Paper IV, birch pollen has also been measured on the roof at Stockholm University. The measurement techniques and the instruments that have been used for the measured pollutants at Torkel Knutssonsgatan in Stockholm are presented in Table 1.
**Table 1. The measurement techniques that have been used for the different air pollutants.**

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Method</th>
<th>Instrument</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO\textsubscript{x}</td>
<td>Chemiluminescence</td>
<td>AC32M, Environnement S.A., France</td>
</tr>
<tr>
<td>NO\textsubscript{2}</td>
<td>Chemiluminescence</td>
<td>AC32M, Environnement S.A., France</td>
</tr>
<tr>
<td>O\textsubscript{3}</td>
<td>UV absorption</td>
<td>O342M, Environnement S.A., France</td>
</tr>
<tr>
<td>PM\textsubscript{10}</td>
<td>Gravimetric</td>
<td>TEOM 1400A, Thermo Fisher Scientific, USA</td>
</tr>
<tr>
<td>PM\textsubscript{2.5}</td>
<td>Gravimetric</td>
<td>TEOM 1400A, Thermo Fisher Scientific, USA</td>
</tr>
<tr>
<td>BC</td>
<td>Transmission of light through a filter</td>
<td>Aethalometers 8100, AE31, AE33 Magee Scientific Corporation, USA</td>
</tr>
<tr>
<td>PNC\textsubscript{7} and PNC\textsubscript{4}</td>
<td>Optical counting</td>
<td>Condensation particle counter, Model 3752/3022 TSI incorporated, USA</td>
</tr>
<tr>
<td>Birch pollen</td>
<td>Manual counting in microscope (Soldevilla et al. 2007)</td>
<td>Burkard Seven Day Recording Volumetric Spore Trap (Burkard Manufacturing Co. Ltd. UK)</td>
</tr>
</tbody>
</table>

**Chemiluminescence** has been used to measure NO\textsubscript{x} and NO\textsubscript{2}. This technique is based on a reaction in a chamber between NO and O\textsubscript{3}, of which energy is transmitted in the form of emitted photons. The emitted photons are then detected by a photo-multiplier tube, in which the output voltage is proportional to the NO concentration.

**UV absorption**, that has been used to measure O\textsubscript{3}, is based on measuring the UV light absorption by ozone molecules. The O\textsubscript{3} concentration is estimated by using two signals, where one of the signals passes directly through a measurement cell, while the other passes through a scrubber used as a catalytic converter, which completely eliminates the ozone molecule. A ratio between these two signals is obtained, where adjustments for pressure and temperature finally give the O\textsubscript{3} concentration.

The **gravimetric** measurement technique, that has been used to measure PM\textsubscript{2.5} and PM\textsubscript{10}, is based on a method called tapered element oscillating microbalance (TEOM), where a hollow glass tube is used as a microbalance. The inlet only allows particles with a special cut-off size (usually PM\textsubscript{2.5} or PM\textsubscript{10}) to enter the tube. The particles entering the nozzle are deposited on a filter, and the change in mass due to the particle absorption creates a change in the oscillation frequency of the filter container. The change in frequency is converted into an electrical signal, which gives a measure of the particle mass that is collected on the filter.

The **transmission** method has been used to measure BC. This technique is using an aethalometer, which means that the concentration of optically absorbing dark suspended particles from a gas stream are measured. The gas stream flows through a filter where the particles are trapped. A light beam is projected through the deposited material, where the dark particles absorb and attenuate this light. The increase in attenuation between two
measurements is proportional to the increase in density of the absorbing material that has been suspended on the filter. This increase in density is proportional to the particle concentration in the gas stream that has been sampled. 

**Optical counting** has been used to measure particle number count ($\text{PN}_{4}$ and $\text{PN}_{7}$). $\text{PN}_{4}$ and $\text{PN}_{7}$ means the particle number count of particles with an aerodynamic diameter larger than 4nm and 7nm, respectively. A condensation particle counter (CPC) has been used, and it works in a first step by enlarging the particles by using them as nucleation centres in order to create droplets in a supersaturated gas. The nucleation process is initiated by heating the inflowing air, in order to obtain a high vapour content. In the next step, the air enters a cooler, where the new particles attach to the droplets. The temperature difference between the heater and the cooler determines the particle size that can be detected, where a relatively larger temperature difference means that smaller particles can be detected. The particles then flow into a reservoir, where a laser counter measures the particle number by laser pulses scattered by the particles.

**Manual counting of birch pollen in a microscope** is based on the Burkard Seven Day Recording Volumetric Spore Trap. This spore trap is equipped with a drum, where a clockwork mechanism moves the drum at a constant rate past a narrow slit in the casing which encloses the drum. A vacuum pump is pumping in air through this slit at a constant rate. Inside this drum, there is an adhesive coated transparent tape consisting of a sticky cellulose strip, where the pollen grains pumped in get stuck. The collected pollen grains are then counted manually in a microscope.

### 4. Summary of results

#### 4.1 Paper I: The use of carbonaceous particle exposure metrics in health impact calculations

**4.1.1. Soot particles and their properties**

The main objective of Paper I is that from a health perspective analyse the different measurement techniques that have been used during the history to measure carbonaceous particles. Carbonaceous particles, colloquially called soot particles, arise as a by-product from incomplete combustion of liquid or gaseous fuels. Even if adequate oxygen is supplied, fuel-rich, oxygen-poor zones can occur when the reactants are not well mixed, which benefits the formation of soot particles (Bond et al., 2013). These particles are small spherical elementary carbonaceous particles clustering with each other. When the combustion products cool down, organic compounds are attached and accumulated to the elementary carbon core. So, in reality, soot particles consist of a mixture of elemental carbon (EC) and organic carbon (OC) (Seinfeld and Pandis, 2006). Depending on the combustion material and the environmental conditions, other materials such as oxygen, nitrates, sulphates and metals are also incorporated onto the particles (Chang et al., 1982). A summary of the properties of soot particles is shown in Table 2, and Figure 3 shows the structure of a soot particle.
Table 2. A description of the properties of soot particles and their formation mechanisms according to Bond et al. (2013) and Seinfeld and Pandis (2006).

<table>
<thead>
<tr>
<th>Property</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arising from combustion processes and they are formed primarily in flames</td>
<td></td>
</tr>
<tr>
<td>They exist as aggregates of small carbon spheres</td>
<td></td>
</tr>
<tr>
<td>They strongly absorb light with a mass absorption cross section of at least 5 m²g⁻¹</td>
<td></td>
</tr>
<tr>
<td>They are insoluble in water</td>
<td></td>
</tr>
<tr>
<td>They are refractory with a vaporisation temperature close to 4000K</td>
<td></td>
</tr>
<tr>
<td>They have a density of around 2 g cm⁻³</td>
<td></td>
</tr>
<tr>
<td>The ratio of carbon and oxygen in the combustion process is largely responsible for their formation</td>
<td></td>
</tr>
<tr>
<td>High combustion temperatures tend to reduce the appearance of soot</td>
<td></td>
</tr>
<tr>
<td>Also associated with OC, metals and secondary materials, depending on the combustion material and the environmental conditions</td>
<td></td>
</tr>
</tbody>
</table>

Figure 3. A soot particle and its structure. Initially, the particle is formed as a primary aerosol consisting of an elemental carbon core, which is caused by incomplete combustion. The particle is then growing larger by gas-to-particle conversion processes, where other components, like organic carbon, sulphate, nitrate and metals are attached to the elemental carbon core, forming a secondary aerosol (Seinfeld and Pandis, 2006).

The different measurement techniques that have been used as a base for health impact calculations are black smoke (BS), black carbon (BC), absorption (Abs) and elemental carbon (EC). They are all measuring airborne carbonaceous particles, but the methods are dependent on the aerosols’ optical, thermal and physicochemical characteristics, and the results may differ substantially (Lavanchy et al., 1999). In the case of estimating health effects associated with exposure to combustion particles, the use of the different measurement methods makes it difficult to obtain consistent and reliable results. Figure 4 describes the two basic principles (light-absorption classification and thermal-optical classification) that are used in order to measure soot particles.
Figure 4. The two basic principles (black carbon and elemental carbon) that are used to measure soot particles, where light absorption is used for BC and a thermal-optical method is used to measure EC. The measurement technique-specific split point is the point where the carbonaceous material is assumed to represent the pure EC fraction (Hagler, 2015).

4.1.2. Description of the different measurement methods (Abs, BC, BS, EC and SC)

The oldest standardized test method is black smoke (BS), which dates back until the 1920s. Originally, it was developed to measure an index called Black Smoke Index (BSI), which was based on a comparison of a soot blemish on a filter with the mass of the collected particles (PM) (OECD, 1964). The BS measurement method is uncertain because the conversion of the soot blemish into PM has remained the same, while the quantity and the chemical composition of the PM have changed significantly, especially as the coal burning in the cities has declined (Quincey, 2007). In other words, when the black-smoke calibration was introduced, it corresponded well to the total mass concentration of the PM sample, but the changes in PM composition have led to that this relationship is no longer valid.

The principle of the elemental carbon method (EC) is thermal conversion of the carbon content of the particles into CO$_2$, which is converted into CH$_4$ for analysis in a flame ionization detector (Birch and Cary, 1996). This method is based on that the carbonaceous material is sampled on a quartz-fibre filter and then heated in an inert helium atmosphere. The heating process will remove the organic carbon (OC), in order to create a pure soot fraction. However, the heating process can cause pyrolysation or charring of the OC, and that may overestimate the mass of EC. However, by simultaneous measurements of the reflectance or transmittance, this source of error can be reduced (Janssen et al., 2012). The main problem associated with this method is that there have been no standardized guidelines in order to carry out these measurements; different laboratories are using different temperature protocols, which will
have a major impact on the results. The consequence arising from this is that the relation between EC and the total carbon can vary by a factor up to five (Cavalli et al., 2010). The process of measuring EC is described in Figure 5.

**Figure 5.** The process of measuring EC (from left to the right in the figure). First, in an oxygen-free helium atmosphere, the sample is heated in order to remove the OC. The pyrolytic carbon (PC) is monitored by measuring the transmission of a laser through the filter. The helium atmosphere is then switched to an oxygen/helium mixture, where the EC is oxidized to CO$_2$ (Maenhaut, 2012).

The term **black carbon (BC)** refers to the measurement of carbon particles by light absorption (transmission). The collected particles give rise to a blemish on the surface of the filter, and a specific absorption coefficient is used to convert the absorption to a mass concentration ($\mu$g m$^{-3}$). BC is a mixture of EC and the light-absorbing OC compounds (Schaap et al., 2007). Usually, EC and BC correlate well, but the influence from the light-absorbing OC compounds constitutes a source of error when the purpose is to measure elemental carbon. In addition to OC, wind-blown dust, iron oxides from railways and non-exhaust emissions from brakes and tires can contribute to this light absorption, which also constitutes a source of error for this measurement technique (Janssen et al., 2012). The principle of measuring BC is described in Figure 6.
The black-carbon measurement technique is based on a filter, where soot particles are collected. A light beam passes through the filter with the collected particles, and the light attenuation ($I/I_0$), caused by the light absorption of the collected soot particles, is converted into a mass concentration, measured in $\mu g m^{-3}$.

The absorption method (Abs) is very similar to the BS measurement technique described above. It is based on measurements of reflectance on a filter with sampled particles using a particle diameter cut-off of 2.5 $\mu m$ (PM$_{2.5}$). Conversion of the results into a mass concentration is usually not performed, which is the rule of the BS measurement technique. However, the Abs values can be transformed into BS values.

The term soot carbon (SC) is another type of measurement for combustion particles. This method is used to distinguish between soot particles originating from fossil fuels from those originating from biomass burning. It is based on an isotope analysis where $^{14}C$ quantification is conducted with a chemo-thermal oxidation method (Andersson et al., 2010)

**4.1.3. Uncertainties associated with the measurement techniques**

The measurement techniques involve particles extracted from air onto a filter, and the measuring of light absorbance or light reflectance. If not performed correctly, erroneous and variable results may be recorded because the particles’ properties change when they are extracted onto a filter (Watson et al., 2005). The most important factors that may cause variable and uncertain results are according to Fuller et al. (1999) the following:

1) Absorption and scattering depend on wavelength, and this is also dependent on particle characteristics that may vary in time and space;

2) Different shapes of the particles affect the optical properties;

3) Variable refractive index;

\[
Tr = I/I_0
\]
4) Inaccurate densities, where different types of particle agglomerates can give the same extinction effects, while the density varies. This results in uncertainties when the light extinction is translated into particulate mass; and

5) Impurities, where the graphitic material is mixed with other substances causing different optical properties of the particles.

The implication for epidemiological studies is that it is difficult to compare different studies, e.g. the relative risks associated with different health outcomes, since the exposure metrics differ between studies. Even if studies use the same indicator, e.g. BS, the results may be affected by spatially and temporally variable optical properties of the particles leading to uncertainties in exposure estimates. The concept RR (relative risk) means the ratio of the probability of an outcome in an exposed group to the probability of an outcome in an unexposed group.

4.1.4. Relationships between BS, BC, EC and Abs

In order to be able to compare the RRs associated with exposure to carbonaceous particles from different epidemiological studies, it is necessary to use the same metric. Considering the health effects associated with exposure to the EC fraction, the RRs will of course vary depending on the size of the calculated EC value. A review of the published studies presenting results of comparisons of the different metrics of “soot” (BS, BC, EC and Abs) is conducted in this section. EC is here considered as the “reference” metric to which the other metrics can be related. But it should be kept in mind that none of the metrics are absolute due to the lack of a traceable standard (as discussed above). In the report from WHO (Janssen et al., 2012), they use the relation EC = 0.11 BS, which is based on a mean value calculated from a variety of different relations. In Table 3, the relationships between EC and BS from different studies are shown. The studies represent different geographical locations from different countries during different time periods.
Table 3. Percent EC content in BS in the different studies. The largest value is 26 % and the smallest 5 %, meaning that it differs by a factor of just over 5.

<table>
<thead>
<tr>
<th>Country</th>
<th>Geographic location and reference</th>
<th>Time period</th>
<th>Percentage EC content in BS</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Rural, Urban, traffic</td>
<td>1999–2000</td>
<td>13 %</td>
<td>Cyrys et al., 2003</td>
</tr>
<tr>
<td></td>
<td>Rotterdam</td>
<td>2006–2007</td>
<td>9 %</td>
<td>Keuken et al., 2011</td>
</tr>
<tr>
<td></td>
<td>Urban</td>
<td>1998–1999</td>
<td>9 %</td>
<td>Schaap et al., 2007</td>
</tr>
<tr>
<td></td>
<td>Rural</td>
<td>2001–2002</td>
<td>6 %</td>
<td>Schaap et al., 2007</td>
</tr>
<tr>
<td>Germany</td>
<td>Berlin; urban</td>
<td>1989–1990</td>
<td>18 %</td>
<td>Erdman et al., 2003</td>
</tr>
<tr>
<td></td>
<td>Munich; urban, traffic</td>
<td>1999–2000</td>
<td>16 %</td>
<td>Cyrys et al., 2003</td>
</tr>
<tr>
<td>USA</td>
<td>Washington; urban, traffic</td>
<td>No info.</td>
<td>15 %</td>
<td>Edwards et al., 1983</td>
</tr>
<tr>
<td></td>
<td>New York; urban, traffic</td>
<td>1996</td>
<td>8 %</td>
<td>Kinney et al., 1996</td>
</tr>
<tr>
<td></td>
<td>New York; urban, traffic</td>
<td>1999</td>
<td>5 %</td>
<td>Lena et al., 2002</td>
</tr>
<tr>
<td>Sweden</td>
<td>Stockholm; urban, traffic</td>
<td>1996</td>
<td>9 %</td>
<td>Hansson et al., 1997</td>
</tr>
<tr>
<td></td>
<td>Stockholm; rural, urban, traffic</td>
<td>1999–2000</td>
<td>7 %</td>
<td>Cyrys et al., 2003</td>
</tr>
<tr>
<td></td>
<td>Stockholm; urban, traffic</td>
<td>1996</td>
<td>6 %</td>
<td>Hansson et al., 1997</td>
</tr>
<tr>
<td>UK</td>
<td>London; urban, traffic</td>
<td>1999–2000</td>
<td>12 %</td>
<td>Adams et al., 2002</td>
</tr>
<tr>
<td></td>
<td>UK (Birmingham, Edinburgh, Halifax and London)</td>
<td>2008–2009</td>
<td>26 %</td>
<td>Heal et al., 2012</td>
</tr>
<tr>
<td>Mean value of all relations</td>
<td>*</td>
<td>*</td>
<td>12 %</td>
<td>*</td>
</tr>
</tbody>
</table>
Figure 7 shows EC values as a function of BS values at different locations in the Netherlands. In Figure 7, it is clear that the percentage EC content in BS is larger at urban locations in comparison to rural locations. Figure 8 shows EC as a function of black smoke, measured as British Smoke Shade at an urban and a traffic site in Washington State. In Figure 8, there are no clear differences between the percentage EC contents in BS at the urban site in comparison with the traffic site. Figure 9 shows BC values as a function of black smoke values at different locations in Great Britain. The data points in Figure 9 show a large spread, but without any clear differences between the different locations. Table 3 shows the large variations in the percentage EC content in BS from different studies at different locations during different time periods. Figure 7–9 show that there are also large corresponding variations within the countries, and also at the specific measurement locations.

Figure 7. Elemental carbon as a function of black smoke at different rural and urban locations in the Netherlands (Schaap and Denier van der Gon, 2007).
Figure 8. Elemental carbon (µg m\(^{-2}\)) as a function of black smoke, measured as British Smoke Shade, BSS (µg m\(^{-2}\)). The measurements have been performed in Washington State. The dashed lines represent the uncertainties as a 95 % confidence interval (Edwards et al., 1983).

Figure 9. Black carbon as a function of black smoke at different locations in Great Britain (Heal and Quincey, 2012)
4.1.5. Comparing health risks associated with exposure to BS and PM$_{10}$

Considering the large differences between the EC content in BS at different places and times, it is difficult to obtain a uniform link between the different exposure measures, which both are intended to measure soot particles. From a health perspective, it is consequently also difficult to get a unified relationship between the health impacts associated with exposure to BS, and the health impacts associated with exposure to EC.

To illustrate the large range that might appear when the health risks associated with exposure to Abs or BC are compared with the health risks associated with exposure to PM$_{10}$, the concentrations of Abs or BC and PM$_{10}$ have been compiled at several different urban sites in Europe (Putaud et al., 2004; Reche et al., 2011; Eeftens et al., 2012). The mortality risks associated with BS have thereafter been calculated based on the proportion of the EC content in BS obtained from Table 3, where the average EC content is estimated to represent a proportion of 12 % in BS (minimum 5.2 % and maximum 27 %). This means that the mortality risk associated with exposure to BS potentially can vary by a factor up to five, depending on the ratio between EC and BS. In comparison with the mortality risk associated with PM$_{10}$, the calculated risk increase for EC can be either significantly larger or significantly smaller, depending on the relation between BS and EC, and also depending on the different relationships between the concentration of EC and PM$_{10}$ in the different cities. The relationships are illustrated in Figure 10.

![Figure 10](image_url)

**Figure 10.** The ratio of relative increase in all-cause mortality associated with short-term exposure to measured levels of PM$_{10}$ vs. exposure to the calculated concentrations of EC (Table 3), where the average EC content is estimated to represent a proportion of 12 % in BS (minimum 5.2 % and maximum 27 %).
In addition to the large uncertainties associated with the transformation from one soot measurement into another, the lack of standardized guidelines for EC measurements makes the situation even more complicated when health risks associated with soot particles shall be assessed and evaluated. An overall conclusion of this study is that a more uniform measurement technique is required to overcome all those uncertainties.

4.2. Paper II: Trends in air pollutants and health impacts in three Swedish cities over the past three decades

Paper II analyses the trends in air pollution concentrations in Stockholm, Gothenburg and Malmo during the period 1990–2015. The main objective of this study was to analyse the health impacts that arise as a result of increasing or decreasing air pollution trends. A centrally located measuring station in each city, representing urban background levels, has been used to measure NOx, NO2, O3, and PM10 in Stockholm, Gothenburg and Malmo during the period 1990–2015. The Openair package (Carslaw and Ropkins, 2012) has been used to obtain a trend for each pollutant in each city. Figure 11 shows the trends for each pollutant in Stockholm performed in Openair, divided into different percentiles.

Figure 11. Trends in NOx, NO2, O3 and PM10 at an urban background site in Stockholm during the period of 1990–2015. The trends are divided into different percentiles, with concentrations representing the 90th percentile (red), the 50th percentile (blue) and the 10th percentile (green).
The average change in $\mu g m^{-3} year^{-1}$ for each pollutant in each city has been used in order to calculate the change in health impact during the period 1990–2015. To quantify the change in health impact associated with change in exposure, the change in life expectancy arising from these changes has been calculated in AirQ+ (WHO, 2016) according to a log-linear exposure-response function. The measured concentrations and trends obtained from the measuring stations may, however, not be representative for the entire population living in those cities. The spatial variation in air pollution concentrations, and the population density within the cities, are important to take into consideration in order to calculate population-weighted exposure concentrations. This is assessed by comparing model-calculated annual population-weighted exposure concentrations with annual-mean urban-background concentrations. This has been done for NOx in Stockholm and Gothenburg, and for O3 and PM$_{10}$ in Stockholm, but due to the lack of model-calculated annual population-weighted concentrations for all cities and pollutants, these relations have not been possible to calculate other than for the above-mentioned pollutants and cities. For NOx and PM$_{10}$, the average exposure concentrations for the urban population are lower compared to the measured urban-background concentrations. The reason for this is that the concentrations are higher at the urban background site in comparison with the concentrations in the outskirts of the urban area. In contrast to NOx and PM$_{10}$, the average exposure concentrations associated with O3 are higher compared to the measured urban-background concentrations. This can be explained by the interaction between NOx and O3, where the O3 reaction with NO dominates over the formation of new O3, and this reaction is more apparent in the central part of a city, where the NO concentration is relatively higher.

With change in life expectancy as a measure of the health impact associated with change in exposure, an interesting question is how much of the total life expectancy increase can be attributed to decreased air pollution exposure. The life expectancy has increased by between four and six years in Stockholm, Gothenburg and Malmo during the period 1990–2015 (SCB, 2017). The change in life expectancy associated with the changing trends in all pollutants in the three cities is presented in Table 4. For NOx and NO$_2$, there are decreasing trends for all the three cities, which means that the exposures to these pollutants have decreased during this period, which have given rise to increases in life expectancy. The reverse applies to O3, where increasing trends in all cities have given rise to decreases in life expectancy. For PM$_{10}$, the trends are less clear and consistent with a less clear impact on the change in life expectancy.
Table 4. Change in life expectancy in months with 95 % CI in brackets, caused by change in exposure during the measured periods. Decreasing trends are associated with an increase in life expectancy, and increasing trends are associated with a decrease in life expectancy (minus signs). The change in life expectancy, adjusted for population-weighted exposure concentrations, are presented in bold below the ordinary values. Note that the trend for PM$_{10}$ in Stockholm is only for the period 1997–2015.

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Stockholm</th>
<th>Gothenburg</th>
<th>Malmo</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO$_x$</td>
<td>21 (11–32)</td>
<td>17 (9–26)</td>
<td>13 (7–19)</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>13 (6–21)</td>
<td>6 (3–9)</td>
<td>7 (3–11)</td>
</tr>
<tr>
<td>O$_3$</td>
<td>-1 (-0.5 – -2)</td>
<td>-2 (-1 – -4)</td>
<td>-2 (-1 – -4)</td>
</tr>
<tr>
<td>PM$_{10}$</td>
<td>0.6 (0–1)</td>
<td>-1 (0 – -2)</td>
<td>No significant change</td>
</tr>
<tr>
<td></td>
<td>0.2 (0.0–0.5)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

When the change in life expectancy associated with the trends (Table 4) is compared with the total change in life expectancy by between four and six years, the change in NO$_x$ is estimated to contribute by up to 20 % of this increase, which may be considered a very large effect. However, the increasing trends associated with O$_3$ give rise to a loss in life expectancy, but this effect is very small in comparison with the gain in life expectancy associated with the decreasing NO$_x$ trends. For PM$_{10}$, the effect on the life expectancy is very small, and the trends for PM$_{10}$ do not exhibit the statistical power as do the trends for NO$_x$, NO$_2$ and O$_3$. To sum up, NO$_x$ and NO$_2$ are indicators of combustion-related air pollutants in general, and an overall conclusion of the study is that reductions of these pollutants may have a great potential to benefit the public health.


In this study, the effects on daily mortality in Stockholm associated with short-term exposure to ultrafine particles (measured as number of particles with a diameter larger than 4 nm, PNC$_4$), black carbon (BC) and coarse particles (PM$_{2.5-10}$) have been compared with the effects from more common traffic-pollution indicators (PM$_{10}$, PM$_{2.5}$ and NO$_2$) and O$_3$ during the period 2000–2016. The associations between different air pollutants and the daily mortality were modelled by using a quasi-Poisson regression model, while controlling for other time-varying factors that can affect daily mortality. The purpose of this study was to compare the different results when estimating the health effects on daily mortality based on different air pollution indicators.

The different air pollutants that were measured exhibited clear seasonal variations. The mean concentrations of BC, NO$_2$ and PNC$_4$ exhibited the highest concentrations during winter (October–March) and lowest in the summer (June–July). Contrariwise, PM$_{10}$, PM$_{2.5}$, PM$_{2.5-10}$ and O$_3$ exhibited peak concentrations during late winter to early summer (March–May). The
reason for these seasonal patterns is largely dependent on the sources of the different pollutants. NO\textsubscript{2}, \textit{PNC\textsubscript{4}} and to a large degree BC are mainly influenced by local vehicle exhaust emissions. The peak during wintertime associated with these exhaust-related pollutants is caused by a more intensive traffic, a more stable atmosphere and lower wind speeds. In contrary, during the summer months, the exhaust-related emissions are lower because the traffic intensity decreases, and the meteorological conditions contribute to greater dilution effects. PM\textsubscript{10}, PM\textsubscript{2.5} and PM\textsubscript{2.5-10} originate from both local sources and long-distance transport. The peaks in concentration associated with PM\textsubscript{2.5-10} and PM\textsubscript{10}, and to some extent also for PM\textsubscript{2.5}, during late March and April are caused by the suspension of road dust particles (> 1 µm in diameter) that occurs during late winter and spring in Stockholm, when the road surfaces become drier (Johansson et al., 2007; Denby et al., 2013). For O\textsubscript{3}, the peak in concentration during spring is caused by a combination of long-distance transport of global tropospheric ozone, and by a large-scale peak in the local photochemical production (Monks, 2000).

The health effects in terms of excess risks associated with daily mortality were calculated for each pollutant during 2000–2016. The excess risk for each pollutant was calculated in a single-pollutant model (Figure 12), and traffic-related pollutants with excess risks with significance levels where the p-values were smaller than 0.2 were included in multi-pollutant models together with O\textsubscript{3}. The excess risks in the single-pollutant model were calculated for both lag01 and lag 02, which means the excess daily mortality associated with the pollutant concentration of the mean of the same day and the previous day, and the mean of the same day and the previous two days, respectively. Pollutants that were negatively correlated, or positively correlated, but with opposite effects in the single-pollutant model were also included in the multi-pollutant models. The significantly positive excess risk associated with O\textsubscript{3} remained significant in two-pollutant models after adjustments for PM\textsubscript{2.5-10}, BC and NO\textsubscript{2}. The significantly positive excess risk associated with PM\textsubscript{2.5-10} remained significant in a two-pollutant model after adjustment for NO\textsubscript{2}. The significantly negative excess risks associated with NO\textsubscript{2} remained significant in two-pollutant models after adjustments for PM\textsubscript{2.5-10}, O\textsubscript{3} and BC.
In Figure 12, there are significantly positive excess risks associated with the non-exhaust related pollutants (O₃, PM₂.₅₋₁₀ and PM₁₀), while the exhaust-related pollutants (PM₂.₅, PNC₄, NO₂ and BC) are not positively significant (p-value < 0.05 is considered as statistically significant). However, the excess risks associated NO₂ are instead significantly negative. First of all, the possibility to quantify any associations between mortality and different pollutants depends on how well the exposure can be quantified. In this study, the measurements from one urban-background measuring site has been used, assuming that the temporal variability at this site reflects the temporal variability of the exposure in the population. Small spatial variations within a city mean that one centrally located measuring station should be enough when it comes to estimating the population exposure. In Stockholm, PM₂.₅ is mainly influenced by non-local sources, and the temporal variations will therefore be very similar everywhere in the city (Cyrys et al., 2003). The O₃ concentrations in Stockholm are also largely influenced by long-distance transport, even though the titration by NOₓ to some extent has an impact on the O₃ concentrations. Pollutants mainly caused by long-distance transport also exhibit relatively small spatial variations within the city. For the exhaust-related pollutants (PNC₄, BC and NO₂), where road traffic is an important source, the spatial variability within a city is larger. Despite these spatial variations, the temporal variations within a city are expected to be quite similar, as traffic intensities usually show similar temporal variations along different roads in a city.

In time-series studies, spatial variations are of limited importance as long as the temporal variations exhibit similar patterns within a city. Despite possible differences in the temporal variations associated with PNC₄, BC, NO₂, it is probably not the main reason for the lack of significantly positive risk coefficients (Figure 12). The reasons for these results can instead be explained by behavioural factors. The results in Figure 12 exhibit a tangible pattern, with significantly positive excess risks for O₃, PM₂.₅₋₁₀ and PM₁₀ (lag02), but with non-significant excess risks for all the exhaust-related pollutants, except for NO₂, which instead exhibits...
significantly negative excess risks. There are some possible reasons for the robust associations between exposure to O\textsubscript{3}, PM\textsubscript{2.5-10} and PM\textsubscript{10}, and the daily mortality. One possible reason may be that the measurement data for O\textsubscript{3}, PM\textsubscript{2.5-10} and PM\textsubscript{10} reflect the exposure better than for the exhaust-related pollutants due to behavioural factors. Increased O\textsubscript{3} concentrations coincides with sunny and warm days in spring and summer when people spend more time outdoors and allow windows to be opened to a greater extent. This will increase the exposure and thereby contribute to the significant excess risks. The concentrations of PM\textsubscript{2.5-10} and PM\textsubscript{10} also tend to increase during sunny days in spring and early summer due to suspension from dry roadways, which will then cause a higher exposure during these days. Contrariwise, the exhaust-related emissions tend to be highest during wintertime when the outdoor activities tend to be lowest, which possibly can contribute to the absence of significant positive associations for all exhaust-related pollutants. The significantly negative excess risks associated with NO\textsubscript{2}, which also remain significant in the multi-pollutant models after adjustments for O\textsubscript{3} and PM\textsubscript{2.5-10}, can be caused by misclassification in exposure due to the use of only one centrally located measuring station.

The main conclusion of this study is that the exhaust-related pollutants exhibit much more uncertain relationships with human mortality in comparison with O\textsubscript{3}, PM\textsubscript{2.5-10} and PM\textsubscript{10}. The difference in exposure due to behavioural factors, where the highest concentrations of O\textsubscript{3}, PM\textsubscript{2.5-10} and PM\textsubscript{10} coincide with that part of the year with more pronounced outdoor activities, can probably largely explain the significant excess risk associated with these pollutants. In the same way but vice versa, the exhaust-related pollutants tend to be highest when the outdoor activities are least pronounced, which probably contributes to the lack of positive significant excess risks associated with these pollutants.


In Paper IV, an air quality health index (AQHI) has been calculated for Stockholm based on data during the period 2015–2017. Short-term health effects in terms of asthma emergency department visits (AEDV) associated with exposure to NO\textsubscript{x}, O\textsubscript{3}, PM\textsubscript{10} and birch pollen have been used as metrics. The main purpose of this study was to calculate the combined health risks caused by simultaneous exposure to several different air pollutants. With four exposures, which together constitute an air quality health index (AQHI), it is possible to calculate both their combined effect, and also the relative importance of each individual air pollutant during different time periods.

In order to calculate an AQHI, the beta-coefficients for Stockholm (Table 5) have been developed. The beta-coefficient (\(\beta\)) represents a standardized coefficient showing how many standard deviations a dependent variable will change, per standard deviation increase in the predictor variable. The beta-coefficients in Stockholm have been calculated by analysing data regarding the daily number of asthma emergency department visits in Greater Stockholm (population 1.2 million) from January 2001 until December 2005. Data were analysed by using additive Poisson regression models for overdispersed counts. The three air pollutants and birch pollen and meteorological variables were included as mean of the same day and the day before (lag01), with adjustments for possible confounders. The averaging exposure-time of 24
hours has been used, and all age groups have been included. The daily mean values of the concentrations of NO$_x$, O$_3$ and PM$_{10}$ have been measured at an urban background monitoring site at Södermalm in Stockholm, while the levels of birch pollen have been measured on the roof of the Arrhenius Laboratories building at Stockholm University. The percentage increases in respiratory hospital admissions during 2015–2017 are presented in Figure 13, where the contribution from each pollutant and their annual variations also are presented.

The beta-coefficients for Stockholm are also compared with the beta-coefficients obtained from a meta-analyses, which are also presented in Table 5. The beta-coefficient (β) represents the linear relationship between the health impact and the change in exposure. For Stockholm, the beta-coefficients associated with exposure to PM$_{10}$ and birch pollen are statistically significant, while the coefficients associated with exposure to NO$_x$ and O$_3$ are not. In the meta-analysis, the coefficients for NO$_2$ and PM$_{10}$ are significant, while the coefficient for O$_3$ is not. The differences between the beta-coefficients in Stockholm and in the meta-analysis can have several different explanations. In comparison with the meta-analysis, the coefficients in Stockholm are based on relatively small amount of data, which can contribute to the lack of significant coefficients for NO$_x$ and O$_3$. The meta-analysis is largely based on studies from the 90’s, while the coefficients in Stockholm are based on data obtained during the period 2001–2005. This can also contribute to the difference between the beta-coefficients in Stockholm and in the meta-analysis. Despite the fact that NO$_x$ is used in Stockholm, while NO$_2$ is used in the meta-analysis, the mean value of the coefficient in the meta-analysis is almost three times larger compared to the mean value of the coefficient in Stockholm. Both NO$_x$ and NO$_2$ are indicators of exhaust emissions from traffic. The use of catalysts and particle filters, which gradually have begun to be used during the 1990’s, may have some impact on the difference between the NO$_2$ coefficient obtained in the meta-analysis during the 90’s and the NO$_x$ coefficient obtained in Stockholm during the period 2001–2005.

Table 5. The beta-coefficients in percent for asthma emergency department visits associated with $10\,\mu\text{g m}^{-3}$ increase of NO$_x$, O$_3$, and PM$_{10}$ in Stockholm. For birch pollen, the beta-coefficient associated with an increase of ten pollen per cubic metre has been used.

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Stockholm</th>
<th>Meta-analysis (Anderson et al., 2007)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO$_x$</td>
<td>0.5 (95% CI: -1.2–2.2)</td>
<td>Not included</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>Not included</td>
<td>1.4 (95% CI: 0.6–2.2)</td>
</tr>
<tr>
<td>O$_3$</td>
<td>0.3 (95% CI: -1.4–2.0)</td>
<td>1.6 (95% CI: -0.1–3.3)</td>
</tr>
<tr>
<td>PM$_{10}$</td>
<td>2.5 (95% CI: 0.3–4.8)</td>
<td>3.3 (95% CI: 1.0–5.5)</td>
</tr>
<tr>
<td>Birch pollen</td>
<td>0.26 (95% CI: 0.18–0.34)</td>
<td>Not included</td>
</tr>
</tbody>
</table>
The risk increase in percent for asthma emergency department (AEDV) visits associated with exposure to the prevailing air pollutants in Stockholm during 2015–2017.

The risk increases in Figure 13 associated with the four pollutants follow a characteristic pattern, reflecting the annual variations in concentration levels. The risk increase associated with NO\textsubscript{x} is highest during wintertime, and with a decrease during summertime. The reason for this is that the traffic is more intensive during wintertime, in combination with a more stable atmosphere with lower wind speeds. During summertime, the traffic intensity and the NO\textsubscript{x} emissions decrease, and the meteorological conditions also contribute to greater dilution effects. The risk increase associated with O\textsubscript{3} is highest during spring, and that is caused by a combination of long-distance transport of global tropospheric ozone, and by a large-scale peak in the photochemical production (Monks, 2000). The peaks in PM\textsubscript{10} concentration and the corresponding peaks in the risk increase during late winter and early spring are caused by an increase in particles originating from road abrasion. The road abrasion is caused by the use of studded tires during wintertime, and when the roads become drier during late winter and early spring, the suspension of road-dust particles reaches an annual maximum. As much as 90% of the mass of PM\textsubscript{10} in Stockholm during the spring originates from road abrasion (Johansson et al., 2007). The risk increase associated with birch pollen is concentrated to the birch flowering period that occurs during April to June. The biennial pattern with high concentrations during 2016, but with considerably lower concentrations during 2015 and 2017 (Figure 13) is caused by a resource competition between vegetative and reproductive organs of the trees. Every two years, growth is high while reproduction is low and vice versa, as can be seen from this biennial pattern associated with birch pollen (Latalowa et al., 2002; Ranta et al., 2005).
The Air Quality Index (AQI) is commonly used in order to describe the air quality situation in relation to a reference value at a given time and place. This index is usually based on several pollutants, where each pollutant is compared with its air quality limit value, and the pollutant with the highest value in relation to its limit value determines the AQI-value (Hewings, 2001). The AQI is based on a four-point index scale, representing low, moderate, moderately high and high pollutant levels. When comparing the AQI, based on PM$_{10}$ in Stockholm during the period 2015–2017, with the AQHI during the same period, these two indices don’t correlate well. The AQHI sometimes show relatively high values for the risk increase even when the AQI represents low and moderate levels. The main reason why these two indices do not correlate is that the AQI is based on one pollutant (the pollutant with the highest value in relation to its limit value) while the AQHI is based on the sum of the health risks associated with simultaneous exposure to NO$_x$, O$_3$, PM$_{10}$ and birch pollen.

To sum up, the introduction to an AQHI may potentially have several advantages. By shifting from a single-pollutant approach to a multi-pollutant approach, the complexity of exposure and health impact can be characterized in a more complete manner, and identifying the no-threshold concentration-response relationship between air pollution exposure and health may also be easier when several pollutants are taken into consideration simultaneously (Stieb et al., 2008). Above all, public information regarding the health risks associated with different pollutants and birch pollen can be very useful for sensitive persons when planning their outdoor activities.

5. Discussion

In this thesis, where health effects due to air pollution exposure have been analysed based on a number of different aspects, there are several different topics that can be discussed. Below follows a review where each paper is discussed separately, and then finish with a more general discussion.

5.1. Soot particles and health effects

In Paper I, where it is clear that the measurement techniques for soot particles are associated with large uncertainties, an important issue that arises is what consequences this will have with regard to legislation and action strategies. In order to being able to make proper regulatory measures to reduce the adverse health effects caused by airborne particles, it is of great importance to increase the understanding of how they affect human health. For soot particles, the use in risk assessments and regulations is limited, because the measurement methods, and the related results, are associated with these large uncertainties. The importance of soot particles in terms of health impact has, however, been addressed in several studies (e.g. Grahame et al., 2014; Janssen et al., 2011). Being able to regulate the emissions of combustion-related particles is of course difficult considering the lack of reliable results from a uniform measurement technique. However, the current results can be improved if the uncertainties are handled in a better way. Handled in a better way means that the relative risks obtained in epidemiological studies, using different measurement methods, are combined by using intervals for the transformation instead of using fixed values, which at
least illustrate the sensitivity to assumptions. A more uniform measurement technique would also be beneficial to introduce in future research. The need for a more uniform measurement technique regarding EC has also been addressed by The European Committee for Standardization (CEN) Technical Committee 264 on Air Quality. They have produced a standardized method for the measurement of the EC and OC content in PM$_{2.5}$. This standard method is validated by laboratory and field measurements, and through statistical analyses of these. This procedure is described in more detail in Brown et al. (2017).

Despite the large uncertainties associated with the measurement techniques that are intended to measure soot particles, there is also research in the field of toxicology providing support for soot particles as particularly important in terms of harmful health effects (Grahame & Schlesinger, 2010). So far, however, very few studies have been performed in order to distinguish the health effects due to EC (elemental carbon) from those of other constituents of PM$_{2.5}$. However, two studies that have investigated health effects related to exposure to diesel exhaust emissions suggest that EC may not be a directly toxic component of PM$_{2.5}$, but it may act as a carrier of several different chemical constituents of varying toxicity, like PAHs and metals (Lucking et al., 2011; Mills et al., 2011).

In Paper I, the short-term health effects associated with EC exposure have been compared with the corresponding health effects related to exposure to PM$_{10}$ at different sites in Europe. Based on the mean concentrations of EC and PM$_{10}$, the health impacts are quite similar when these two metrics are compared. However, the RRs associated with EC are about 10 times higher compared to PM$_{10}$ per 1 µg m$^{-3}$, but the health effects associated with the mean values are quite similar due to the lower EC concentrations. Regarding long-term effects related to EC and PM$_{10}$ exposure, similar relationships prevail. When the health benefits in terms of gain in life years saved associated with decreased long-term exposure to EC and PM$_{10}$ were calculated in Rotterdam during 1985–2008, the increases were 12 and 13 months per person for EC and PM$_{10}$, respectively, despite that the population-weighted exposure concentrations decreased by 2 µg m$^{-3}$ and 18 µg m$^{-3}$ for EC and PM$_{10}$, respectively (Keuken et al., 2011). According to these results, reductions in the EC emissions are much more effective when it comes to benefit the public health. So, from a policy point of view, soot particles are of particular interest to address when it comes to introducing action strategies in order to reduce the emissions.

5.2. Health impacts related to trends in air pollution concentrations

5.2.1. Explanations for the trends

In Paper II, the presentations of the air pollution trends for NO$_x$, NO$_2$, O$_3$ and PM$_{10}$ give a clear picture of the development during the last three decades. The increase in life expectancy has increased by between four and six years in Stockholm, Gothenburg and Malmo during the period 1990–2015. Given that NO$_x$ is considered as a good indicator of vehicle exhaust, the calculations in Paper II show that up to 20 % of these increases are attributable to the decreased NO$_x$ emissions, which may be considered as a very large contribution. The European emission standards (Euro), which were introduced in the early 90’s, have largely contributed to the decreasing trends associated with NO$_x$ and NO$_2$. The continuous implementation of all stricter regulations regarding emissions through these standards during 1990–2015 has
furthermore contributed to that the trends have continued to decline. In Stockholm, however, the trends associated with NOx and NO2 cease to decline from 2007 onwards. An increased proportion of diesel vehicles can at least partially explain these changes. The lack of the same phenomenon in Gothenburg and Malmö is largely unexplained, but the fact that the proportion of new registrations of diesel vehicles has been greatest in the Stockholm area during this period can possibly explain these slightly increasing NOx and NO2 trends seen in Stockholm (BilSweden, 2018).

Despite that the decreasing NOx trends have had significant positive impacts on public health, it should not be interpreted as the problem of air pollution exposure is not a serious issue any more. The premature deaths related to air pollution exposure are still of a significant magnitude, where the prevailing conditions in Stockholm and Gothenburg are shown in Segersson et al. (2017). The relatively large impact on life expectancy attributed to the decreasing NOx trends should rather be interpreted as if it exists a great potential to benefit the public health by continuing to reduce the traffic emissions. From a policy point of view, the results in Paper II could possibly help to increase the incentives for creating mitigation strategies in order to reduce the air pollution concentrations within the cities.

5.2.2. NOx and NO2 and the health effects associated with exposure

When it comes to estimating the health effects associated with exposure to NOx and NO2, there are several factors that need to be considered. Both NOx and NO2 are indicators of traffic-related emissions in general. To what extent they are harmful in themselves, and to what extent they are indicators of other harmful pollutants, e.g. soot particles and ultrafine particles, is not entirely clear. However, when considering simultaneous exposure to PM2.5 and NO2, the relative risks were not significantly affected when included in a two-pollutant model, indicating that total PM2.5 is not driving the associations with NO2 (Faustini et al., 2014).

In Paper II, the calculated change in life expectancy between 1990 and 2015 is based on studies of associations between mortalities during the 70s and the 80s, i.e. much earlier than the period considered in this paper. An important question to address is if these RRs are valid for this 25 year period. The composition and correlation of the combustion-related traffic emissions in the Swedish cities have changed significantly during the period 1990–2015. Assuming that NO2 and NOx are indicators of exhaust emissions in general, and that the trends associated with exhaust-related PM have decreased differently as a result of the introduction of catalysts and particle filters, then it might be inappropriate to use the same RRs for the entire period from 1990–2015.

Based on emission factors according to HBEFA (Keller et al., 2017), the trend in the emissions of NOx during the period from 1990 up until around 2010 is quite similar to the trend for exhaust-related PM. The trend for PM/NOx in the exhaust of the vehicle fleet in Stockholm is shown in Figure 14. This indicates that NOx and NO2 can be assumed to be representative indicators for exhaust-related combustion particles in general during the first 20 years of the total 25 year period. However, during the period from 2011–2015, the relative amounts of PM to NOx in exhaust have been decreasing.
The reason for these different trends is the introduction of particle filters on newly manufactured vehicles in order to meet the requirements according to the Euro 5, which was introduced in 2011. This indicates that NO\textsubscript{x} is a less appropriate indicator for exhaust-related pollutants in general during the last five years of the measurement period. The consequence of this may be that the calculations of the increase in life expectancy associated with the decreasing NO\textsubscript{x} trends may be underestimated, given that the concentrations of exhaust-related PM have decreased more during the last five years of the total 25 year period. However, since it is only the last part of the period that exhibits diverging trends, and given that the effect on long-term mortality associated with NO\textsubscript{2} is stable after adjustment for PM\textsubscript{2.5} (Faustini et al., 2014), it can be assumed that the life expectancy calculations for NO\textsubscript{x} and NO\textsubscript{2} are not significantly affected by the diverging trends during the last part of the period.

5.2.3. The air pollution trends in other cities in Sweden

In the trend analysis article (Paper II), Stockholm, Gothenburg and Malmo have been analysed with regard to the air pollution trends. An interesting question is if the declining trends regarding exhaust-related pollutants are something that applies to other smaller and less expansive cities in Sweden. In Figure 15, the NO\textsubscript{2} trends in different cities in Sweden are presented as an average change measured as \( \mu g \cdot m^{-3} \cdot year^{-1} \) with 95 % CI. In Figure 16, the black-smoke trends (BS) in different cities in Sweden during wintertime are presented in the same way. The measured trends in the different cities refer to different time periods, but the vast majority refer to the period from early 90s until a period into the 2000s. The weighted trends for both NO\textsubscript{2} and BS exhibit in both cases a statistically significant decreasing trend.
Figure 15. The average annual change in the NO\textsubscript{2} concentrations at different locations in Sweden from the beginning of the 90s until a period into the 2000s. The confidence intervals vary widely, but the weighted overall trend exhibits a statistically significant decline.
Figure 16. The average annual change in BS concentrations during wintertime at different locations in Sweden from the beginning of the 90s until a period into the 2000s. The confidence intervals vary widely, but the weighted overall trend exhibits a statistically significant decline.

In addition to the trends in Stockholm, Gothenburg and Malmo during the past three decades, presented in Paper II, the air pollution concentrations for NO$_2$ and BS exhibit declining trends in most places in Sweden during the same time period. Even though BS has not been measured in Paper II, it is like NO$_2$ and NO$_x$ an indicator of exhaust emissions, except where domestic wood burning occurs, especially in smaller towns. However, since both NO$_2$ and BS exhibit statistically significant overall trends, an increase in life expectancy can be assumed to have occurred for the Swedish populations in general, as a result of the decreasing NO$_2$ and BS trends.
5.3. Behavioural factors as a possible explanation for the results in Paper III

In Paper III, behavioural factors in terms of variations in outdoor activities and ventilation of homes depending on the weather have been proposed as a potential reason for the lack of statistically significant excess risks for the combustion-related pollutants, but where the excess risks associated with \( \text{O}_3 \) and \( \text{PM}_{2.5-10} \) are statistically significant. The importance of different activities depending on season is speculative, and has not really been scientifically verified. However, a similar effect of the influence on exposure misclassification due to behavioural factors has been discussed for \( \text{O}_3 \). When considering \( \text{O}_3 \) exposure, a study performed in the U.S. has analysed the short-term mortality effects associated with \( \text{O}_3 \), and where also temperature and the use of air conditioning were included in the models. They found that higher air conditioning prevalence could significantly mitigate the mortality risk associated with \( \text{O}_3 \) exposure on high temperature days (above the 75th percentile), where the air conditioning prevalence better correlates with the actual use of the equipment, and thus less ozone exposure, especially if also more time is spent indoors (Jhun et al., 2014).

5.4. The air quality health index (AQHI)

Since the air contains a complex mixture of several different air pollutants, it is important to be aware of the additive or possibly also synergistic effects that can arise. The combined exposure, calculated as an air quality health index, may have several advantages over setting the limit value based on the single air pollutant that exhibits the highest concentration in relation to a reference value. By including \( \text{NO}_x \), \( \text{O}_3 \), \( \text{PM}_{10} \) and birch pollen, the AQHI indicates the most important types of pollutants.

In Paper IV, where the AQHI for respiratory emergency hospital admissions has been based on \( \text{NO}_x \), \( \text{O}_3 \), \( \text{PM}_{10} \) and birch pollen, there is an assumption that the total effect is additive; in other words, the AQHI is based on the addition of the individual effect from each pollutant, but without considering possible synergies. However, the evidence of synergism at normally occurring air pollution exposure levels is limited, and in the absence of evidence of synergism or antagonism, the assumption of additive effects may be considered as the default for risk assessments (Mauderly and Samet, 2009). In other words, despite possible interaction effects, an AQHI based on the sum of the individual effect of each pollutant is the method that has the greatest scientific support. However, since the AQHI also includes birch pollen, it is also a factor to take into account. Simultaneous air pollution exposure has been shown to be able to increase the allergic response in a number of studies (Rogerieux et al., 2007; Strand et al., 1998; Svartengren et al., 2000). Since potential interaction effects were not documented, such effects are not now included in the AQHI, but may be in the future if proven.

The beta-coefficients that have been used to calculate an AQHI exhibit very different characteristics regarding the degree of statistical power. The confidence intervals for the coefficients calculated for Stockholm are very large for both \( \text{NO}_x \), \( \text{O}_3 \) and \( \text{PM}_{10} \). For \( \text{NO}_x \) and \( \text{O}_3 \), they are additionally not statistically significant. On the contrary, the coefficient for birch pollen is very clear with a \( p \)-value of \( \text{1.e}^{-10} \). However, the point estimates of the beta-coefficients for the pollutants should also be viewed in the light of results from similar studies.
In the future, it might be possible to calculate local beta-coefficients based on a larger amount of data, and thereby improve the accuracy of this air quality health index.

From a health perspective, the AQHI is assumed to reflect the most important air pollutants. NOₓ is a marker for exhaust, O₃ is a marker for oxidants in the air and PM₁₀ is a marker for road dust. However, an AQHI based on these three air pollutants has its limitations. There is no marker for secondary particles, and not for air pollutants related to wood burning, but in Stockholm, these pollutants could be less important.

5.5. Local risk coefficients or weighted coefficients from a meta-analysis

An important issue to consider is if a city-specific risk coefficient is preferable to a meta-coefficient. It is easy to believe that a coefficient developed in the city that is to be investigated is a better choice than using a meta-coefficient, but the heterogeneity within the city also needs to be considered. City-specific estimates vary around the overall effect estimates due to thru heterogeneity and due to additional stochastic error. There are different potential effect modifiers that can contribute to the heterogeneity (Le Tertre et al., 2005). In Paper II, relative risks obtained from other studies have been used in the health impact calculations, where change in life expectancy has been used as a metric. In the absence of meta-coefficients for NOₓ, the RR of 1.06 (95 % CI 1.03–1.09) per 10 µg m⁻³ increase based on a cohort study in Gothenburg has been used (Stockfelt et al., 2015). A justification for that choice of RR is the similarities between Gothenburg, Stockholm, and Malmö regarding climate, vehicle fleet, and type of city. However, the choice of RR for NO₂ in Paper II is based on a meta-analysis, where random effects estimates were reported, and where the heterogeneity across the analysed studies was assessed by using Chi-squared test and the inconsistency test (I²) (Faustini et al., 2014). The RR of 1.066 (95 % CI 1.029–1.104) per 10 µg m⁻³ increase in NO₂ that has been used in Paper II, based on (Faustini et al., 2014) is, however, very close to the RR of 1.06 (95 % CI 1.03–1.09) per 10 µg m⁻³ increase in NOₓ, based on the cohort study in Gothenburg, that has been used in the calculations in Paper II, even if it is NOₓ that has been used Stockfelt et al. (2015), while NO₂ has been used in Faustini et al. (2014). Considering that NOₓ in the cities in Paper II to between 60 and 80 percent consists of NO₂ during the period 1990–2015, then it can be assumed that the NOₓ coefficient is valid to use in the calculations.

For O₃, there is a limited number of long-term studies associated with mortality. In Paper II, the use of the RR of 1.01 (95 % CI 1.005–1.02) per 10 µg m⁻³ increase is based on a large prospective long-term cohort study performed in the U.S. (Turner et al., 2016). This RR estimate is, however, very close to the RR of 1.014 (95 % CI 1.005–1.024) per 10 µg m⁻³ increase from another large cohort study performed in the U.S. (Jerrett et al., 2009). There are no meta-analyses regarding long-term exposure to O₃ available. But if there are no specific effect modifiers for long-term ozone exposure in Sweden, which are not present in the U.S., the use of these coefficients should be valid also for Sweden.

The RR of 1.04 (95 % CI 1.00–1.09) per 10 µg m⁻³ increase used for the calculations of PM₁₀ in Paper II is based on meta-analysis based on 22 European cohorts (Beelen et al., 2014). The corresponding RR value from the same study associated with exposure to PM₂.5-10 is 1.04 (95 % CI 0.98–1.10) per 10 µg m⁻³ increase. Since PM₁₀ to a large part consists of PM₂.5-10 in the
Swedish cities, the similarity between the RRs for PM$_{10}$ and PM$_{2.5-10}$ can justify the use of this RR in the Swedish cities.

In Paper IV, where the AQHI has been calculated, the local risk coefficients have been used in the calculations. Even with the uncertainties associated with the use of local risk-coefficients (Le Tertre et al., 2005), we have chosen to use local estimates. The reason for this is that the available meta-coefficients are obtained from studies during the 90s, and with many studies from limited parts of the world. Especially with regard to NO$_x$ and NO$_2$, these coefficients may have limited relevance to the current air quality situation, where much has happened in terms of development and use of catalysts and particle filters on newly manufactured vehicles. For O$_3$, the use of the local coefficient can be justified by the adjustment for birch pollen that has been made, which has not been done by most of the studies included in the corresponding meta-coefficient. The coefficients for PM$_{10}$ are surprisingly similar, considering the unusual situation in Stockholm with road dust from studded tires as a significant source to the mass of PM$_{10}$. However, the relatively few studies included in the meta-coefficient for PM$_{10}$, with an over-representation from North America, have made us choose local risk-coefficient obtained in Stockholm. For birch pollen, there are no meta-coefficients available to use.

5.6. Limitations of basing the population exposure on air pollution data from a single urban-background measuring site

The results in Paper II, Paper III and Paper IV are based on measurement data from urban background stations. When health effects related to air pollution exposure are addressed, it is important to be able to estimate the population-weighted exposure. The reliability of using one centrally located measurement station is different depending on whether short-term effects or long-term effects are considered. In Paper III and IV, only the short-term effects are considered. For pollutants with high temporal correlations within a city, one measurement station might be considered as valid enough in order to estimate the short-term effects related to exposure, because the day-to-day variations of the considered pollution concentrations can be expected to be quite similar everywhere in a city, although the total concentrations may be different at different locations. However, even though traffic-related emissions tend to be highly correlated at different parts within a city, as has been shown for PNC in Augsburg by Cyrys et al. (2008), and in Amsterdam, Athens, Birmingham and Helsinki by Puustinen et al. (2007), these results may not be applicable to all cities. Different emission sources and atmospheric processes in different cities, which are not only dependent on traffic emissions, mean that the temporal correlations of PNC at different sites are not high in all cities (Reche et al., 2011). For BC, the spatiotemporal correlations at different measurement sites in Stockholm were not very high (R < 0.70) for the daily average concentrations, as shown in Kreci et al. (2011). The same relationships can be assumed to prevail for NO$_x$ and NO$_2$, where the main source is local emissions from traffic. High correlations between NO$_x$ and BC in Stockholm, with R-values in the range of 0.76–0.90 at three measuring sites, were also found in the same study (Kreci et al., 2011), indicating that there are spatiotemporal variations also for NO$_x$ and NO$_2$. Strong spatial and diurnal variability associated with traffic-related emissions
of PM$_{2.5}$ have also been shown in Toronto, but where no spatial and temporal differences were observed for the non-traffic-related contributors of PM$_{2.5}$ (Jeong et al., 2019).

The use of studded tires in Stockholm means that the coarse fraction of PM$_{10}$ (PM$_{2.5-10}$) largely consists of road dust. Importantly, meteorology can have a significant impact on the concentrations of PM$_{10}$ through both surface and dispersion conditions (Norman et al., 2016). The spatial variations in road dust are probably large because different roads have varying proportions of vehicles with studded tires, different speeds, different coatings and different amount of sanding and salting. The temporal variations in road dust are controlled by time variations in meteorology and time variations in traffic flows of light and heavy vehicles. The meteorology affects the wetness of the roads, which may vary depending on whether the roads are sunlit or in shade. The wetness of the roads has proved to be an important factor in terms of the emissions of road dust (Johansson et al., 2007).

These above mentioned spatiotemporal variations in the air pollutant concentrations create uncertainties in time-series studies, when it comes to estimating population exposure based on one centrally located measuring station.

5.7. Long-term mortality effects compared to short-term effects

When the health effects related to air pollution exposure are addressed, the increased mortality is often used as a metric. In both Paper I, Paper II and Paper III, mortality has been used as a metric for the health effects. An important thing to be aware of is that the mortality caused by air pollutants is not evenly distributed among the population. In time-series studies, like Paper III, the number of deaths related to acute effects associated with air pollution exposure are calculated, but the loss of life expectancy (LLE) is not shown. In other words, it is not known to what extent the air pollution attributable deaths have shortened the lifetime for the affected individuals. However, a reasonable assumption is that the LLE per death is in average very small (in the order of months) in comparison with e.g. traffic accidents with estimated corresponding mean values of 30–40 years (Rabl, 2003). So, in terms of social benefits, it is important to be aware of these differences when evaluating the benefits of avoidance related to different factors contributing to premature deaths. However, the long-term effects related to air pollution exposure are also an important factor. In long-term cohort studies, it is possible to calculate the total LLE based on the change in age-specific mortality associated with long-term exposure (Rabl, 2003). In Paper II, the change in life expectancy associated with air pollution trends has been calculated based on risk coefficients for long-term effects, where up to 20 % of the increase in life expectancy during 1990–2015 was attributed to decreasing NO$_x$ trends. To sum up, even if the LLE per death in time-series studies is assumed to be relatively small, the long-term effects in terms of increased life expectancy associated with decreased exposure are large when NO$_x$ and NO$_2$ are used as indicators.
5.8. Mortality compared to other health-related indicators

When comparing air pollution attributed mortality with other indicators in short-term studies, e.g. hospital admissions and emergency department visits, mortality can be considered as the most extreme outcome, and where less serious outcomes are not noticed. However, in comparison with other health outcomes, e.g. emergency department visits, mortality is a more reliable indicator. The reason for this is that emergency department visits and hospital admissions as indicators exhibit an uncertainty because different individuals are differently inclined to seek medical care for air pollution attributed health problems, which means that unrecorded statistics may occur when these kind of indicators are used. Different access to health care in different countries and cities also creates uncertainties when it comes to comparing results that have been obtained at locations where the access to health care is significantly different. In other words, the number of emergency department visits tends to be higher in a city with relatively good access to healthcare compared to a city with relatively poor access to healthcare, although the health-related problems with air pollution exposure would be equal. On the contrary, mortality as an indicator does not give rise to the above-mentioned problems with unrecorded statistics, which makes it a more accurate and comparable indicator. However, the age structure within a population can affect the results in time-series studies, where a population with a larger proportion of older people gives a relatively greater impact on the daily mortality at a certain air pollution concentration.

When using mortality and change in life expectancy as metrics in order to assess the health effects related to air pollution exposure, it is important to be aware of that it is a limitation and underestimation of the total impact. Cardiovascular and respiratory morbidity are not illustrated, although these effects occur before mortality, which may be considered as the top of the iceberg. In Paper II, where the change in life expectancy is calculated as a result of increasing or decreasing air pollution trends, several other effects not related to mortality can also be assumed to occur. The most obvious decreasing trends for NOx in Paper II, with a relatively large subsequent effect on the life expectancy increase, are also expected to reduce the effects on cardiovascular and respiratory morbidity (Johansson et al., 2009). However, there are also other systemic effects associated with air pollution exposure, many of which are poorly represented by using mortality as an indicator. Recent years of research also highlight the impact of air pollution exposure on metabolic diseases, pregnancy and developmental outcomes, the central nerve system and on psychiatric disorders (Thurston et al., 2017). In Paper II, it is shown that the decrease in life expectancy associated with the increasing O3 trends is small in comparison with the increase in life expectancy associated with decreasing NOx trends. However, when using pregnancy outcomes as a metric, O3 was associated with an increased risk of pre-eclampsia and preterm birth, while no effects were observed for the other air pollutants (Olsson et al., 2013). Consequently, the increase in life expectancy shown in Paper II, where the effect associated with O3 is small in comparison with NOx, can be completely different if pregnancy outcomes is instead taken into account. To sum up, mortality as an indicator of health effects related to air pollution exposure has its limitations, but also has its strength in terms of more reliable statistics in comparison with many other health outcomes.
6. Conclusions

The uncertainties associated with the measurement techniques for soot particles must be addressed in a different way than has been done before. Since there is strong evidence for combustion-related particles as particularly important in terms of health effects, this is an important issue to consider. Although there are long historical time series of soot measurements from many cities in Europe, the possibility of making health risk analyses based on these is limited, because the chemical composition of the particles exhibits large spatial and temporal variations, and it is therefore not possible to determine universal relationships between the different measurement techniques for soot particles. To be able to make reliable health impact assessments associated with exposure to soot particles, a more uniform measurement technique is urgently needed in order to overcome these uncertainties.

The impact of decreasing air pollution concentrations in Stockholm, Gothenburg and Malmo during 1990–2015 has had a very large effect on the population's expected life expectancy. The trends in air pollutants in Stockholm, Gothenburg and Malmo during 1990–2015 clearly show the great importance of reducing the air pollution concentrations in order to benefit the public health. The relatively large increase in life expectancy attributed to the decreasing NOx trends provides further support for exhaust emissions as particularly important to consider in the case of mitigation strategies and legislation measures.

When comparing the associations between daily mortality and the concentrations of several different air pollutants in Stockholm during the period 2000–2016, the excess risks associated with exposure to exhaust emissions (NOx, PM2.5, BC and PNC4) exhibit much more uncertain relationships in comparison with O3, PM2.5–10 and PM10. The reason for these findings is probably to a large part explained by behavioural factors, where the concentrations of O3 and PM2.5–10 are in general high during sunny days, when outdoor activities tend to increase, while the opposite applies to NOx, PM2.5, PNC4 and BC, with the highest concentrations during the winter months.

The air quality health index (AQHI) is a useful tool when it comes to estimating the health risks associated with simultaneous exposure to several different air pollutants. By shifting from a threshold-based index (AQI) to a health-risk based index (AQHI), based on NOx, O3, PM10 and birch pollen, there are several advantages that can be achieved. Public information regarding the expected health risks associated with current or forecasted concentrations of pollutants and pollen can be very useful for sensitive persons when planning their outdoor activities. For interventions, it can also be important to know the contribution from pollen and the specific air pollutants, judged to cause the risk. The relative importance of each pollutant during different parts of the year is also illustrated through this multi-pollutant approach.

7. Outlook

All the uncertainties related to the measurements of combustion-related particles and their health impacts entail some limitations in terms of introducing regulatory measures. Continued research in this area is needed in order to improve the understanding of the causal relationships of how particle exposure affects the health. The uncertainties associated with the measurement techniques are probably easier to overcome by means of introducing a
more uniform method of measurement. An overall conclusion from Paper I is that a more specific and standardized methodology for measuring combustion-related particles is urgently needed. This would lead to more reliable results in the future, when health impacts are calculated in epidemiological studies. This could also make it easier when it comes to implementing a robust and standardized measurement technique for soot particles in the legislation.

Soot particles originating from biomass burning are something that may be important to consider in the future. Since the use of biofuels can be expected to increase as a result of climate awareness, the findings in Paper I are important to pay attention to as exposure to soot particles can be expected to constitute a serious health issue even in the future. Continued research regarding the biological mechanisms associated with exposure to soot particles is needed in order to clarify what makes them harmful to the health. To distinguish between traffic generated EC and biomass generated EC may also be important in order to discover possible differences in their toxicity. A better understanding of all these biological mechanisms can potentially make it easier to develop the most suitable action strategies.

The trend analyses in Paper II show the importance of continued work within this research field. General awareness regarding trends of different air pollutants can have several advantages. In addition to the public health perspective, the trends can possibly also be used in order to evaluate the importance of different types of mitigation strategies.

The concept AQHI, described in Paper IV, may have several advantages when it comes to being used as a warning and monitoring system. First of all, all the included pollutants are addressed simultaneously, which reflects the actual situation in a more satisfactory manner. An AQHI based on asthma emergency department visits can also be used in order to provide warnings regarding the air quality for today, or for the coming days, with a weighted index. A geographical representation with a map, where it is shown how the index varies over different parts of a city, can also be helpful for people to be able to make conscious choices in order to avoid places that exhibit a relatively high index value. This can be particularly important for children and elderly, and for those who have health problems that make them particularly sensitive to air pollution exposures. The AQHI in Paper IV is based only on asthma emergency department visits (AEDV). In the future, the design of this index could possibly be improved. The AQHI could also be based on mortality risks or other disease outcomes, e.g. hospital admissions for cardiovascular diseases. Maybe it would be possible to use several weighted outcomes in order to include as many air pollution-related health effects as possible. However, the issue regarding possible interaction effects resulting from combined exposure to several different air pollutants, where both synergistic and antagonistic effects can occur, requires further research.

8. Acknowledgements

First of all, I want to thank my two supervisors Christer Johansson and Bertil Forsberg for guiding me true this long and laborious time as a PhD student. A special big thanks to Christer Johansson who has been the principal supervisor, and who has always been very supportive throughout the whole process. Since this PhD thesis has not been linked to any predetermined project, I am convinced that this has made it extra tough for the supervisors. In addition to
being supervisors, they also have had to define relevant scientific issues, and how these should be addressed, which they have managed to do very well. I also want to give a special thanks to Johan Ström for giving me the opportunity to conduct a postgraduate education at ACES, which I am very grateful for. A special thanks to Hans Orru who has given me major support during parts of this thesis. Great thanks also to Christofer Åström, Kadri Meister and Andreas Tornevi for the statistical support they have provided me with. And finally, a great thanks to Kevin Noone for reviewing this thesis before printing.

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