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The influence of atmospheric circulation and meteorology on urban air pollution and pollen exposure

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Front page photo: Urban area of Gothenburg, Sweden. Photo taken by Håkan Pleijel.

Abstract

Urban air quality is a global health concern and is a growing problem due to large migration of people from rural areas to cities, a phenomenon occurring in many parts of the world. This means that more and more people can be expected to be exposed to high levels of air pollutants, many of which are associated with the urban environment. The exposure situation is characterised by different compounds emitted from different sources such as traffic, industry, wood burning and energy production. Air pollution levels tend to vary temporally both during the day and between seasons. Another important atmospheric constituent to consider is pollen which together with air pollutants can cause severe health effects in sensitive people. The climate and weather governs the atmospheric processes responsible for ventilation and stagnation of the air, which in turn also provides conditions for good or poor air quality. This thesis has investigated the urban air pollution levels of nitrogen oxides ($NO_x = NO + NO_2$), ozone (O_3), particles (PM_{10} and PNC, particle number concentration) and birch pollen levels in relation to meteorology and atmospheric circulation. In this study circulation was represented by the large scale circulation pattern called the North Atlantic Oscillation (NAO) and by the synoptic circulation classification scheme Lamb Weather Types (LWT). The city of Gothenburg has been the main location but air quality and pollen in Malmö has also been investigated. It was shown that air pollution has a strong association to the variation in weather conditions represented by both NAO and LWTs. In winter calm and stagnant air masses were associated with high levels of NO and NO_2, these conditions were more common NAO was in its so called negative mode (characterized e.g. by low wind speeds) and in LWTs associated with calm conditions and thus limited ventilation. Ultrafine particles (UFP), considered to be of large importance for health effects, are in many cases the dominating fraction in PNC. NO_x was found to be a good proxy of PNC, e.g. situations with high NO_x can be expected to have high PNC. Furthermore, the occurrence of high NO_2, O_3 and PM_{10} were co-varying very well with the occurrence of high birch pollen counts in Gothenburg. These situations were also associated with high sales of over-the-counter (OTC) antihistamines, indicating a combined effect on health symptoms represented by OTC sales, especially during calm and dry weather conditions. Finally, the usefulness of LWTs was illustrated by to their strong association with anomalies of inter-annual air pollution levels. By adjusting annual concentration/deposition trends of air pollutants for the yearly LWT variability, temporal trends were greatly improved, e.g. the relative importance of weather was quantified permitting more accurate evaluation of emission changes on air pollution levels. Furthermore, the strong association between urban air quality and atmospheric circulation shown in this thesis highlights the LWT classification as a good option to be integrated in a tool for risk assessment and information system for urban air quality including both air pollutants and pollen.

Keywords: Urban air pollution, nitrogen dioxide, particles, ozone, birch pollen, air quality standards, atmospheric circulation, synoptic weather, Lamb Weather Types, North Atlantic Oscillation, meteorology, wind speed, temperature inversions, anomalies.

Populärvetenskaplig sammanfattning

Dålig luftkvalitet är ett globalt och växande hälsoproblem på grund av stora och i vissa fall ökande utsläpp och inflyttning av människor till storstadsregioner, som pågår i stora delar av världen. Exponeringen består av många olika luftföroreningar på grund av ett flertal olika utsläppskällor som exempelvis trafik, industri, vedeldning och energiproduktion. Halten av luftföroreningar varierar i tid, både under dygnet och mellan olika årstider. Pollenförekomst är en ytterligare aspekt att beakta i riskanalyser av den urbana luftkvaliteten. Vissa typer av pollen utgör ett stort hälsoproblem genom att framkalla allergier. Även pollen har en stark säsongsvariation som är kopplad till vegetationens årliga cykel. Klimatet och vädret styr de atmosfäriska processer som skapar förutsättningarna för god eller dålig luftkvalitet, inte minst genom att förstärka eller försvaga den luftomblandning som gör att föroreningarna späds ut. Dålig luftkvalitet uppträder ofta vid låga vindhastigheter som gör att föroreningar ventileras bort mycket sakta.

Denna avhandling har undersökt förekomsten av luftföroreningar; kväveoxider ($NO_X = NO + NO_2$), ozon (O_3), partiklar (PM_{10}) och björkpollen i förhållande till meteorologi och den atmosfäriska cirkulationen framförallt i Göteborg, men delvis också i Malmö. Den atmosfäriska cirkulationen representerades av det storskaliga cirkulationsmönstret, den Nordatlantiska Oscillationen (NAO). Under vintertid har detta mönster en stark koppling till det rådande vädret i Europa. Höga halter av NO och NO_2 i Göteborg var starkt kopplade till kalla och stabila luftmassor, vanligt förekommande under så kallad negativ NAO, vilket betyder att den västliga vinden som vanligtvis råder över Nordatlanten var försvagad. Detta gör att luftmassor från polarområden eller Sibiren kan röra sig in över Sverige.

Förekomsten av höga luftföroreningshalter var även starkt kopplade till väderleken på mindre skala, den så kallade synoptiska vädersituationen, representerad av Lambs vädertyper (LWT = Lamb Weather Types). Situationer med höga halter av kväveoxider visade sig koppla starkt till höga partikelantal i luften, speciellt under vissa LWT. Partikelantal domineras ofta av de minsta partiklarna som kallas för ultrafina partiklar. Dessa tillhör troligen den farligaste kategorin av luftföroreningar när det gäller effekter på hälsan. Vidare visade det sig att halter av björkpollen tillsammans med luftföroreningar ofta var höga samtidigt, speciellt under vissa väderförhållanden. Många av dessa situationer var också tydligt kopplade till förhöjd försäljning av receptfria antihistaminläkemedel, vilket är en indikation på en kombinerad effekt av pollen och luftföroreningar på allergiska symptom.

Slutligen kvantifierades vädrets bidrag till mellanårsvariationen av halter och deposition av luftföroreningar. Det visade sig att mellanårsvariationen i vädertyper till stor del kunde förklara mellanårsvariationen i förekomst av luftföroreningar. Genom att kompensera för den del av mellanårsvariationen som beror på väder får man möjlighet att bättre bedöma hur stor andel av variationen i belastningen av luftföroreningar som förklaras av förändrade emissionsmönster. Detta är viktigt för att korrekt kunna utvärdera effekten av exempelvis strategier för minskade utsläpp på luftkvaliteten.

De resultat och metoder som tagits fram genom denna avhandling kan användas för att förbättra prognosinstrument, riskbedömning och information till allmänheten vad gäller luftföroreningar och pollen.

The influence of atmospheric circulation and meteorology on air pollution and pollen exposure

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This thesis is based on the following papers, referred to in the text by their respective Roman numerals.

I. **Grundström M**, Linderholm H. W., Klingberg J, Pleijel H (2011). *Urban NO_2 and NO pollution in relation to the North Atlantic Oscillation NAO.* Atmospheric Environment 45, 883-888

II. **Grundström M**, Tang L, Hallquist M, Nguyen H, Chen D, and Pleijel H (2015). *Influence of atmospheric circulation patterns on urban air quality during the winter.* Atmospheric Pollution Research, 6, 278-285

III. **Grundström M**, Hallquist M, Hak C, Chen D, and Pleijel H. *Variation and co-variation of PM_{10}, particle number concentration, NO_x and NO_2 in the urban air– relationship with wind speed, vertical temperature gradient and weather type.* (under revision, after review process in Atmospheric Environment)

IV. **Grundström M**, Dahl Å, Ou T, Chen D, and Pleijel H. *The relationship between pollen, air pollution and weather types in two Swedish cities* (manuscript)

V. Pleijel H, **Grundström M**, Pihl Karlsson G, Karlsson P.E, Chen D. *A method to assess the inter-annual weather-dependent variability in air pollution concentration and deposition based on weather typing* (manuscript)

The papers are appended in the end of the thesis and are reproduced with the kind permission from the respective journals.

Scientific papers co-authored by Maria Grundström, which are not included in this thesis:

Grundström M and Pleijel H (2014). *Limited effect of urban tree vegetation on* NO_2 *and* O_3 *concentrations near a traffic route.* Environmental Pollution 189:73-76

Coria J, Bonilla J, **Grundström M**, Pleijel H (2015). *Air pollution dynamics and the need for temporally differentiated road pricing.* Transportation Research Part A, 75, 178-195

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Abbreviations

A	Anticyclone
AQS	Air quality standard
C	Cyclone
Lapse rate	Change in temperature with altitude
LWT	Lamb Weather Types
MSLP	Mean sea level pressure
NAO	North Atlantic Oscillation
NAOI	North Atlantic Oscillation Index
NO	Nitrogen oxide
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides ($NO + NO_2$)
O_3	Ozone
OTC	Over-the-counter
PBL	Planetary boundary layer
PM	Particulate matter
PM_{10}	Particulate matter with an aerodynamic diameter < 10 µm
P	Atmospheric pressure
PNC	Particle number concentration
ppb	Parts per billion
RH	Relative humidity
T	Air temperature
u	wind speed
UFP	Ultrafine particles
VOC	Volatile organic compound
VPD	Vapour pressure deficit
ΔT	Temperature difference between two heights

1. Introduction

Every day we inhale approximately 10-25 m^3 of air containing to a large part of N_2 (78.08%), O_2 (21.90%) and argon (0.93%), but also trace compounds such as suspended particles, nitrogen oxides and ozone, all known to have negative health effects. Many large urban areas around the world suffer from episodes of very poor air quality, and an estimated 3.7 million deaths on a global scale has been attributed to ambient air pollution where south-east Asia is largely affected (WHO, 2014). Small urban areas like Gothenburg, in south-west Sweden also experience days with high air pollution levels. The severity of poor air quality is largely connected to emission sources and the prevailing weather conditions. Furthermore, air pollution during the pollen seasons add further health risks for sensitive people. Air pollution and pollen may have interactive effects.

1.1 Air pollutants and birch pollen

1.1.1 Nitrogen oxides

Nitrogen gas (N_2) is the most abundant pure element on earth, comprising approximately 78% of Earth's atmosphere. The reaction between N_2 and oxygen (O_2) produces nitrogen oxides ($NO_x = NO + NO_2$) and requires very high temperatures. In nature, such temperatures are provided by lightning and natural forest fires. Anthropogenic sources are predominantly related to combustion in vehicle engines, industrial processes and wood burning. The high temperature in vehicle combustion engines causes the N_2 in the air to react with O_2. In many urban environments the dominant source of NO_x emissions is road traffic and a large fraction is emitted as NO, varying between 80-90% depending on types of vehicles (Alvarez et al., 2008; Carslaw et al., 2011), the rest is emitted as NO_2. NO and NO_2 participate in many chemical reactions involving titration with ozone and photochemical dissociation, further described in section 1.1.3. NO_2 is removed from the atmosphere through deposition and several reactions involving the OH radical, producing nitric acid (HNO_3), a sticky molecule which reacts easily with surfaces, especially wet surfaces (Finlayson and Pitts, 2000) where it dissolves into ion forms H^+ and NO_3^-.

1.1.2 Particulate matter

Particulate matter (PM) is a complex mixture of different sized particles containing a broad range of materials emanating from many sources, such as coal power plants, diesel exhausts, sea spray, bioaerosols from wood burning, dust from soil, roads, deserts and volcanoes (Curtis et al., 2006; Gustafsson and Franzén, 2001; Pey et al., 2009). The PM is either directly emitted from the source or produced from co-emitted gases by gas to particle conversion including the production of new particles by nucleation. The composition is related to its source and may compose of trace metals, inorganic salts, low volatile organic compounds or soot. In addition, particles can undergo many different transformations in the atmosphere due to evaporation, condensation and coagulation (Kumar et al., 2011) the relative importance of which is strongly connected to the physical property of the atmosphere e.g. temperature, humidity, solar radiation and wind (Charron and Harrison, 2003; Ketzel et al., 2003; Kumar et al., 2011). These processes essentially change the original composition and size of the particle and add uncertainty to its source identification. PM can be classified according to its diameter size; coarse (PM_{10} < 10µm), fine ($PM_{2.5}$ < 2.5µm) and ultrafine particles (UFP < 0.1µm). High PM_{10} levels in the urban environment are largely related to mechanically generated PM from road transport i.e. clutches, breaks and re-suspension of dust from road and tyre wear (Johansson et al., 2007; Ketzel et al., 2007). The dominant source for UFPs is also related to road transport but their generation occurs in the fuel combustion process and are emitted from vehicle exhausts (Pey et al., 2009; Woo et al., 2010; Kumar et al., 2011). Furthermore, UFPs can be represented by particle number concentration (PNC)

1.1.3 Ozone

The ozone layer in the stratosphere protects life on earth by filtering out some of the dangerous UV-light (UV-C and parts of UV-B). In the troposphere and especially at ground level, however, it is a toxic compound both to humans and vegetation. O_3 is a secondary pollutant meaning it is not directly emitted but rather formed from the reaction of other compounds. It is both produced and destructed in a complex series of solar radiation dependant photochemical reactions involving NO_x, volatile organic compounds (VOCs) and sunlight.

Short-waved sunlight (wavelength < 420 nm) can photolyse the NO_2 molecule, producing atomic oxygen (O), which in turn reacts with molecular oxygen (O_2) to produce O_3:

$$NO_2 + h\nu \xrightarrow{\lambda < 410\ nm} NO + O \qquad \text{Reaction 1}$$

$$O_2 + O \rightarrow O_3 \qquad \text{Reaction 2}$$

Once formed, O_3 may be destroyed from the reaction with NO:

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad \text{Reaction 3}$$

The reaction of O_3 with NO is important near roads with dense traffic and large urban areas where large emissions of NO_x are common producing a net destruction of O_3. A net production of ozone is achieved through VOCs reacting with NO, essentially preventing the destruction of O_3 (Reaction 3) and producing more NO_2 which can form further O_3:

$$RO_2\cdot + NO \rightarrow RO\cdot + NO_2 \qquad \text{Reaction 4}$$

In the urban environment ozone levels tend to be lower than in rural areas due to the titration with NO (Reaction 3). Episodes of high ozone levels in Sweden are often coupled to transport from continental Europe (Solberg et al., 2005).

1.1.4 Birch pollen

Birches (*Betula* L.) are common in the northern temperate and boreal zones of the Northern Hemisphere. They are pioneer species, early to establish in primary successions, and colonize open ground after a disturbance or when pastures and agricultural land are abandoned during a change of land use, as was common i North Europe during the 20th century. The flowers are monoecious and wind-pollinated, flowering before or when the leaves come out. The male flowers with 3 stamens each, are situated, 3 and 3 together, in pendulous ’’catkins’’; each catkin contains 2-300 flowers and may produce 5 million pollen grains (Dahl and Fredrikson, 1996). Pollen grains are haploid male individuals, produced in the microsporangia of seed plants. In flowering plants, the pollen grains contain three cells, two small sperm cells and a large vegetative cell, which is rich in cytoplasm, starch or oil. The vegetative cell produces a wall called the intine, consisting mostly of carbohydrates, and which during development is covered by of sporopollenin, an extremely resistant, complex and elastic biopolymer which is perforated by numerous micropores, allowing for transport of water and soluble compounds. The sporopollenin layer is called the exine. When a pollen grain absorbs moisture on the stigma of a female flower, germination of a pollen tube begins. This tube penetrates the stigma surface and grows between cells through the style towards the ovary. In birches, where pollination is pollen-limited, few pollen accumulate on each stigma during anthesis, and the tubes make an halt at the bottom of the style. When anthesis is over, the tubes make a simultaneous start into the ovary, and compete to be first to enter an ovule (Dahl and Fredrikson, 1996). The two gametes, together with the nucleus of the vegetative cell are transported through the tube to this ovule, and fertilization can take place.

The intensity of the flowering varies between seasons and is highly connected to the previous years´ flowering intensity (Dahl and Strandhede, 1996) and heat accumulation during the period of catkin development (Dahl et al., 2013; Kwarahm et al., 2014). In South Sweden, locally produced birch pollen normally appears in the middle or end of April, and in North Sweden, in the middle or end of May.

1.1.5 Health effects of air pollutants and pollen

Health effects from air pollutants are closely related to the respiratory and cardio vascular systems, but cancer and neurological, reproductive and developmental effects have been associated with exposure to air pollution (Curtis et al., 2006). The exposure takes place through the inhalation of polluted air where the upper parts of the respiratory system efficiently filters out coarse particles. Gases are not filtered out and smaller, especially nano-sized particles reach deep into the lung and can transfer into the blood stream via the respiratory system (Heal et al., 2012). The toxicity of particles is determined by its composition and can carry and deposit toxic substances into the lung. Diesel exhausts, composed to a large degree of UFPs have been determined cancerous by the WHO (IARC, 2012). Seaton and Dennekamp (2003), proposed UFP exposure, proxied by NO_2 to be the cause of cardiovascular effects. Reactive species such as NO_2 and O_3 react with the protective lining inside the lung, slowly breaking down its tissue, known as oxidative stress (Traidl and Hoffman, 2012).

Airborne birch pollen is among the most common sources to pollen allergy in N Europe (de Weger et al. 2013). In order to have a substantial impact to public health in this context, a plant has to be common, it must produce a lot of pollen, the pollen grains have to be wind-borne, and they must contain proteins with allergenic properties, usually called allergens (Frenz 2001). Birches fulfil all these conditions. The most important allergen in birch, Bet v 1 leaks out in large amounts within one minute in moisture (Belin and Rowley. 1971). When an allergy towards a specific allergen is developed, it is called sensitisation, and requires exposure to this allergen. Other environmental factors, such as long- term exposure to air pollutants, may promote the process (Penard-Morand, et al. 2010). Except for allergenic proteins, the pollen grains also leak proinflammatory substances, and enzymes that stimulate oxidative stress (Gilles et al., 2009). Several studies show the connection between pollen counts and symptoms (e.g. Kiotseridis et al., 2013). ”Free” allergens may also occur in the air after leaking out in rain and dew, in airborne debris from broken pollen grains, and secondarily attached to inorganic airborne particles (Schäppi et al., 1997; Taylor et al., 2007; Behrendt and Ring, 2012).

1.2 Atmospheric processes influence ambient levels of air pollution and pollen

1.2.1 Thermal turbulence and atmospheric stability

The fate of suspended air pollutants and pollen is to a large degree determined by the physical properties of the atmosphere e.g. the prevailing weather conditions. Most weather phenomena (e.g. clouds, precipitation, cyclones and storms) occur in the troposphere, and many relevant meteorological processes important to air pollution are active in the lowest layer of the atmosphere close to the earth’s surface known as the planetary boundary layer (PBL).

The vertical mixing is often strong within this layer but fluctuates on a daily and seasonal basis mainly due to variations in heating and cooling of the surface. When short waved solar radiation reaches the Earth's surface it's converted into long-waved thermal radiation, leading to an increase in temperature of ground level air parcels. The air expands, attains a lower density and starts to ascend and is replaced by colder denser air from higher elevation. These conditions are known as neutral atmospheric conditions and the temperature decreases with altitude, on average by approx. 1°C per 100 m increase in height. This is mainly caused by the adiabatic cooling of rising air, meaning the cooling is a result of the decrease in air pressure with altitude without any heat exchange with the surrounding air.

The PBL height normally peaks during the day and is the lowest at late night. At the poles the troposphere reaches an altitude of approximately 10 km above the surface and 15 km at the equator. During high-pressure situations with clear sky and strong solar radiation large vertical eddies are produced causing strong thermal turbulence. This enhances the positive lapse rate (unstable atmosphere) resulting in strong vertical transport of heat, moisture and other compounds such as pollen and air pollutants from the surface up to higher levels of the troposphere (Stull, 1988). At night when solar heating ends and radiative cooling of the surface begins a stable surface layer forms, reducing vertical air mixing (stable atmosphere). During cloudy conditions cooling and heating is reduced, due to thermal radiation being trapped near the surface and weaker solar radiation. This tends to produce a neutral atmosphere, where vertical mixing is neither enhanced nor suppressed.

Under certain conditions the normally occurring temperature decline with height can be inverted (negative lapse rate) when the air layers closest to the ground are colder than layers above i.e. temperature inversions. This is caused by radiative cooling of the ground, which is promoted by a clear sky in combination low wind speeds. Vertical air mixing is suppressed by inversions forming a lid of warm air above cold air, emissions at ground level are trapped near the surface and air pollution levels rise. In Gothenburg, strong ground level inversions typically develop in winter during cold, calm and clear days normally associated with high pressure weather systems (e.g. Olofson et al., 2009).

1.2.2 Advection

Pollutants and pollen are transported horizontally by the wind, a process known as advection. The life time of specific compounds determines the distance which they may travel. Pollen, particles and ozone, have relatively long life-times (~tens of hours to a couple of days for pollen and particles and weeks to months for ozone) and can therefore be transported and dispersed over large distances and areas, away from the emission sources. Episodes of dust particles from large sources such as the Saharan desert or volcanic eruptions occur from time to time (Ansmann et al., 2012). High pollen and O_3 levels may be the result of long-range transport (Ranta et al., 2006; Skjøth et al., 2007; Tang et al., 2009). Furthermore, intercontinental and vertical transport of ozone has been suggested (Derwent et al., 2004; Stohl et al., 2003). On a local scale strong winds tend to ventilate the air and dilute locally emitted air pollutants and pollen (Jones et al., 2010; Grundström et al., 2015; Khwarahm et al., 2014; **Paper III**).

1.2.3 Removal and transformation

In a turbulent dry PBL not only upward movement takes place, the turbulent flux also moves air towards the surface causing continuous removal of air pollutants to the ground known as dry deposition, and can be defined as the deposition that is independent of precipitation (Seinfeld & Pandis, 2006). Air pollutants reactions with different surfaces, uptake in plants and inhalation by humans are all considered dry deposition processes and their relative strength differs between pollutants. Particles dry deposits to the ground due to gravitation; large particles and pollen have a larger deposition velocity than small particles (Faegri and Iversen, 1989).

Wet deposition is the transfer and removal of gases or particles from the atmosphere to the surface by rain, snow and fog (Finlayson and Pitts, 2000). Particles and pollen are very sensitive to precipitation and both are removed efficiently from the atmosphere through wet deposition. Many chemical transformations take place within rain droplets. Similarly to NO_2, sulphur dioxide (SO_2) is removed through oxidation reactions involving the OH radical producing sulphuric acid (H_2SO_4). SO_2 is an air pollutant emitted mainly from combustion of sulphur rich oil and coal in industrial processes but also motor vehicles. When reacting with water it dissolves into its ions $2H^+$ and SO_4^{2-}. Both HNO_3 and H_2SO_4 cause acidification of soil and deposit mainly through wet deposition, like NH_4^+ which together with HNO_3 contributes to eutrophication.

1.3 Atmospheric circulation

1.3.1 Large scale circulation

Atmospheric processes are largely determined by the weather which in turn is connected to the atmospheric circulation e.g. the large-scale movement of several air masses. An air mass is defined as a volume of air covering several 1000 km^2 exhibiting certain metrological properties, often characterised by temperature and moisture, acquired from the surface below. In the northern hemisphere a circulation pattern called the North Atlantic Oscillation (NAO), influence the direction and strength of the westerly wind across the North Atlantic (Chen and Hellström, 1999; Hurrell et al., 2003;). This air mass movement generates climate variability e.g. variations in temperature, precipitation and storminess from eastern North America into western Eurasia and from the Arctic into the subtropical Atlantic, especially during the boreal winter.

The NAO constitutes two semi-permanent atmospheric pressure-centres, the Icelandic low-pressure and Azoric high-pressure. The difference in pressure between these centres is the driving force of the air mass movement across the Atlantic. A large pressure gradient is the result of a deep low and strong high (positive NAO), resulting in very strong westerlies. These conditions produce a large number of strong low-pressures, bringing mild, wet and windy weather into north-western parts of Europe. The strong Azoric high blocks the turbulent weather from southern Europe, which tends to get dry and cold. When the pressure gradient is weak however, the westerlies are suppressed. A smaller number of low-pressure systems are produced and due to the weaker blocking effect from the Azoric high, mild and wet Atlantic air masses travel on a more southerly route over Europe. Arctic and or Siberian air masses move over north-western Europe bringing cold, dry and stagnant weather to the region in negative NAO conditions.

1.3.2 Synoptic scale circulation

There are two important synoptic scale weather systems; the anticyclone and the cyclone. These can be further divided according to the specific latitude belt in which they form, for example; mid-latitude, subtropical and extra-tropical cyclones and anticyclones. The source region determines the specific air-mass properties and may be classified with further detail as either continental or maritime, the latter generally associated with higher moisture content. Normally a combination of latitude belts and surface type is used to classify an air mass; for example, continental polar (~30° < latitude < ~60°), maritime polar and continental arctic (latitude > 60°). Air masses generated over sea tend to be warm (cool in Boreal summer) and wet; while air masses generated over continents tend to be dryer and cold (warm in Boreal summer). The largest and most numerous synoptic systems are the mid-latitude cyclones and anticyclones, active on latitudes 30°-70° (Arya, 1999). Figure 1 shows an example of the average anticyclonic (A) and cyclonic (C) synoptic situation over Gothenburg during spring months March, April and May 2006-2012.

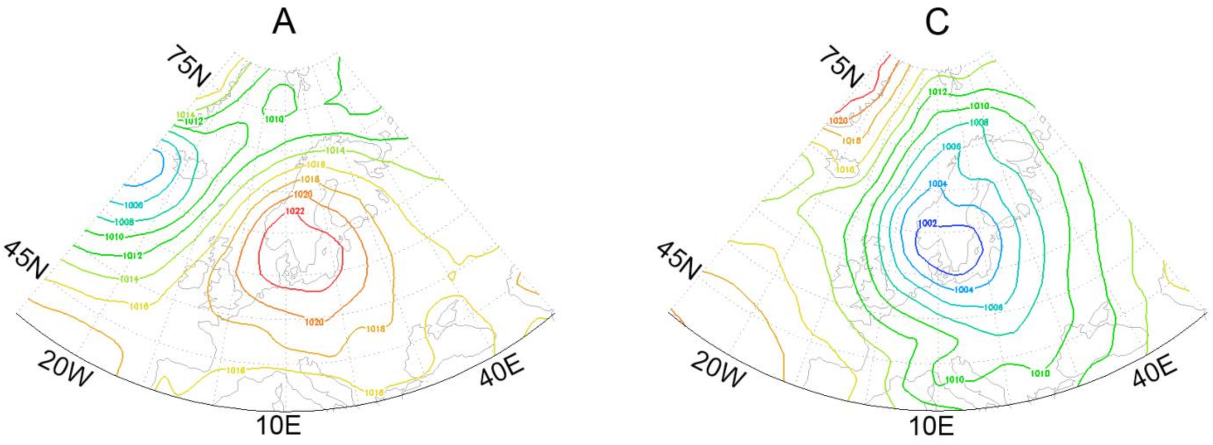

Figure 1. Synoptic composite maps of anticyclonic weather (A) and cyclonic weather (C) cantered over Gothenburg, south-west Sweden. Different colours correspond to different atmospheric pressure, red is high pressure dark blue is low pressure.

The atmosphere flows along the coloured isobars around the pressure centra, clockwise around the anticyclone and counter clockwise around the cyclone. These systems give rise to transport of great amounts of heat and moisture from the tropics to the poles with the southerly air mass flow created in front of the cyclone or behind the anticyclone. The distribution of these systems varies on a day-to-day basis changing the atmospheric directional flow over a specific region. This in turn greatly influence the daily variation in weather conditions. An efficient way to capture the atmospheric vorticity (A or C) or directional flow (N, NE, E...) is to use the objective classification scheme called the Lamb Weather Types. This scheme is based on the variation in the synoptic scale mean sea level pressure (MSLP) and has been proven to be a useful summary of local meteorological conditions (Chen, 2000; Grundström et al., 2015; Tang et al., 2009).

1.4 Regulation of air pollutants

To protect human health and the environment the Swedish government and European Union have enforced air quality standards (AQS), threshold concentration levels for different air pollutants. Each pollutant have specific restrictions on the number of permitted exceedances of threshold levels, based on either yearly, daily or hourly averages. Table 1 illustrates the AQSs for air pollutants relevant in this thesis.

Table 1. Swedish air quality standards relevant in this thesis for NO_2 and PM_{10} and the Swedish environmental objective for O_3.

Time resolution	Pollutant	Concentration threshold	Limit not to be exceeded
Year	NO_2	40 µg m^{-3}	Average **
	PM_{10}	40 µg m^{-3}	Average
Day (24h)	NO_2	60 µg m^{-3}	7 days $year^{-1}$
	PM_{10}	50 µg m^{-3}	35 days $year^{-1}$
	O_3	80 µg m^{-3} *	
Hour	NO_2	90 µg m^{-3}	175 h $year^{-1}$
	NO_2	200 µg m^{-3}	17 h $year^{-1}$ **

* Swedish environmental objective

** EU limit

2. Aims and hypotheses

1. The main and overall aim of this thesis was to assess the variation in urban air quality in Gothenburg during circulation on a large scale (NAO) and synoptic scale (LWTs). In **Paper I**, urban air quality was represented by NO and NO_2 and consideration was taken to the NAO only. In **Paper II**, the same pollutants were investigated with the addition of the influence from LWTs on pollution concentration. **Paper III** included PM_{10}, PNC and **Paper IV** included NO_2, PM_{10}, O_3 and birch pollen and an additional city, Malmö.

2. Air pollution variation is strongly linked to the physical properties of the atmosphere which in turn is linked to the circulation. The second aim was to assess the meteorological character in Gothenburg during large scale (NAO) and regional scale (LWTs) circulation. Since every study covered different time periods, a meteorological characterisation of NAOI (**Paper I, II**) and LWTs (**Paper I, II, III, IV**) was conducted for each study. Paper IV also included the city of Malmö.

3. UFPs are detrimental to human health but lacks ambient air regulation. PM_{10} is dominated to large degree by coarse particles and is therefore a poor estimate of UFPs. The third aim of this thesis was to investigate the relationships between NO_x, NO_2, PM_{10} and PNC and whether a potential proxy relationship was influenced by the variation in meteorological variables and LWTs (**Paper III**).

4. From a health perspective exposure to several toxic compounds can have additive and/or synergistic effects on human health. The fourth aim of this thesis was to investigate the co-variation between air pollutants (NO_2, PM_{10} and O_3) and ambient birch pollen counts during different LWTs. Furthermore it was tested whether simultaneous high levels had an increased effect on symptoms represented by sales of over-the-counter antihistamine drugs (**Paper IV**).

5. There is considerable variation in air pollution levels on an annual time scale which may pose problems when assessing results of emission reductions. The fifth aim was to provide an objective tool to assess the weather influence on annual anomalies in air pollution levels and to improve the possibility to detect temporal trends due to emission changes (**Paper V**).

The specific hypotheses are listed below:

1. Air pollutants reach high levels more often during low wind speed and stable atmospheric stratification due to limited dispersion of local emissions.

2. High levels of birch pollen occur more often during low to moderate winds, low precipitation and high VPD due to limited dispersion and wet deposition.

3. In winter, the occurrence of high NO_2 and NO levels are expected in the negative NAOI and certain LWTs which more often represent anticyclonic vorticity or air mass movement from polar regions in the north or continental land masses in the east.

4. The frequency of LWTs is influenced by the variation in NAOI due to its link with the intensity of the westerly wind across the North Atlantic.

5. NO_x is a good proxy of PNC due to similar emission sources and similar responses to the variation in meteorology and LWTs.

6. In spring, calm and dry LWTs directly link with high levels of air pollutants and birch pollen.

7. A large fraction of the inter-annual variability of air pollution concentrations can be explained by the frequency distribution of LWTs.

3. Material and methods

3.1 Sites and measurements

Gothenburg

Monitoring of air quality and meteorological data was performed on a rooftop 30 m above ground level in the commercial district of Gothenburg city centre (“Femman”; 57°42.52´N, 11° 58.23´E). The site is located adjacent to the central terminal for bus and trains and approximately 300 m away from a busy traffic route (E45). Hourly measurements of NO and NO_2 (Tecan CLD 700 AL chemiluminescence instrument), PM_{10} (Tapered Element Oscillating Microbalance, Series 1400b), PNC (Condensation Particle Counter TSI 3775), atmospheric pressure (Vaisala PA11A), air temperature and relative humidity (Campbell Rotronic MP101 thermometer/hygrometer), wind direction and wind speed (Gill ultrasonic anemometer) were carried out.

Figure 2. Rooftop monitoring station in the commercial district of Gothenburg. Photo taken by Svante Sjöstedt.

The vertical air temperature gradient at two heights (3 m and 73 m above ground) was measured (RM Young platinum temperature probe model 41342) at a site located 8 km south of the city centre (“Järnbrott”; 57° 38,84′N, 11° 55,60′E). The difference in temperature between the two heights (T_{upper}-T_{lower}) represents a measure of the atmospheric stability and is signified by ΔT. This data was used in **Paper II and III**.

Hourly measurements of NO and NO_2 (Differential Optical Absorption Spectroscopy) were performed parallel to a road at ground level in the eastern central part of Gothenburg. That site is located at a busy traffic route (E6/E20) surrounded by low residential buildings, ~5 to 15 m tall (“Gårda”; 57°42.05´N, 11°59.70´E). In **Paper II** data for the winter months (January, February, December) of the period 2001 – 2010 were used for NO and NO_2 from this site.

Birch pollen data was obtained from the Pollen Laboratory, University of Gothenburg for both Gothenburg and Malmö for the years 2006-2012 and was used in **Paper IV**. Pollen data was measured using Burkard Seven-Day Recording Volumetric Spore Traps and provides pollen count with a two hourly resolution. In Gothenburg pollen is monitored on top of rooftop 40 m above ground level at Sahlgrenska University Hospital “Östra” in the eastern part of Gothenburg city (57°43.34´N, 12° 3.12´E). The area is surrounded by residential areas, woodlands in the east and south.

Malmö

In the city centre of Malmö air quality was measured on a rooftop 20 m above ground (“Rådhuset”; 55 36.38’N, 13 0.11’E). Hourly measurements of NO_2 (Eco Physics CLD 700 AL chemiluminescence instrument), PM_{10} (Tapered Element Oscillating Microbalance, Series 1400AB), O_3 (Thermo Enviromental Instruments Model 49C). Approximately 500 meters south of the air quality station (“Heleneholm”; 55 35.21’N, 13 0.61’E), meteorological measurements were carried out.

In Malmö pollen was monitored on a rooftop 30 m above ground at Skåne University Hospital in the southern part of the city centre. The area is mainly surrounded by urban ground with a large park to the west. The region outside of the city is dominated by open agricultural land. Data from Malmö was used in **Paper IV**.

3.2 Lamb Weather Types

Daily mean sea level pressure (MSLP) for a 16 point-grid centred over the Gothenburg city centre (57°7´N, 11°97´E), were obtained from the NCEP/NCAR Reanalysis database 2.5 × 2.5 degree pressure fields (Kalnay et al., 1996). Circulation indices, u (westerly or zonal wind), v (southerly or meridional wind), V (combined wind strength), ξ_u (meridional gradient of u), ξ_v (zonal gradient of v) and ξ (total shear vorticity) describing the geostrophic winds and Lamb weather types (Jenkinson and Collison, 1977) were calculated following Chen (2000). This classification scheme has 26 weather types: anticyclone (A), cyclone (C), eight directional types (NE, E, SE, ...), 16 hybrid types (ANE, AE, ASE, CNE, CE, CSE, ...). In this thesis, the 26 weather types were consolidated into 10 LWTs according to the directions of the geostrophic wind, directional: NE, E, SE, S, SW, W, NW, and N, rotational: A and C (**Paper II-V**).

3.3 NAO index (NAOI)

The NAOI data used in this thesis was obtained from the Climate Research Unit, University of East Anglia; (http://www.cru.uea.ac.uk/~timo/datapages/naoi.htm). The NAOI was calculated on a monthly basis from the difference between the normalised sea level pressure (normalisation period 1951-1980) over Gibraltar and the normalised sea level pressure over Southwest Iceland (e.g. Jones et al., 1997). Monthly index values were used throughout the study. In **Paper I** the time period covered 1997-2006 and the monthly index values varied between -2.25 and 5.26. In **Paper II** the time period covered 2001-2010 and the monthly NAOI varied between -4.85 and 5.26.

3.4 Anomaly analysis

In **Paper V** data for the period 1997-2010 was used for all pollution variables. The annual anomaly z_i of year i of pollutants concentration or deposition (C_i) of year i was defined as (Grumm and Hart, 2001):

$$z_i = \frac{(x_C - C_i)}{\sigma_C} \qquad \text{(eq 1)}$$

where x_c and σ_c is the average and standard deviation, respectively, of the pollutant concentration/deposition during the study period. Linear detrending of the annual data was made before the anomalies were calculated; overarching trends over the period were thus assumed to be the result of changes in emissions, possibly with some influence from long-term climate change and emission changes in far distant sources, especially for tropospheric ozone. The time fraction (f) of each of the ten LWTs was calculated for each year. The estimated annual anomaly caused by variation in the frequency of different LWTs, z_i_LWT, was represented by a linear combination of the time fractions of the different LWTs of year i:

$$z_{i\_LWT} = k_A f_A + k_N f_N + k_{NE} f_{NE} + \ldots + k_{NW} f_{NW} + k_C f_C \qquad \text{(eq 2)}$$

Numerical optimization of the coefficients k_A, k_N, k_{NE} … was made separately for each pollutant index (Excel Solver) to minimize the deviation between observed z_i, and z_{i_LWT} using the least square approach over the study period. Then the observed anomaly z_i was regressed vs. z_{i_LWT} representing the estimated LWT contribution to the anomaly. The relationship was evaluated using linear regression with respect to the coefficient of determination (R^2).

Further, time series of observed annual concentrations/depositions were compared to time series of LWT adjusted annual values. This was made by adding the (positive or negative) z_{i_LWT} to C_i. The observed and LWT adjusted time series were evaluated by R^2, the sum of squares of residuals (SSR, quantifying how much the data deviate from the regression line) and the statistical significance of the regression slope.

Further details about the specific calculations and statistical analysis can be found in the respective papers.

4. Results

4.1 Meteorology governs air pollutants and pollen levels

Local meteorological variables and air pollutants were correlated in Gothenburg. Concentrations of NO_x, NO_2, PM_{10} and PNC were strongly connected to the variation in wind speed and vertical temperature gradient **(Paper III)**. Figure 3 shows an example of averages of NO_x (a) and PNC (b) in relation to intervals of wind speed. Strong (R^2=0.99) negative non-linear relationships were observed for both pollutants. It is clear that the highest levels were observed for wind speeds lower than 2 m s^{-1} and that pollution decreased sharply for wind speeds between 0 – 2 m s^{-1}. This illustrates the importance for calm conditions as a prerequisite for high pollution levels. Similar response patterns in relation to the variation in wind speed but with different magnitudes was valid for NO_2 and PM_{10} (**Paper III, Figure 2b and d**). During conditions with stronger ventilation (wind speed > 3 m s^{-1}) levels were generally very low illustrating the important diluting effect on concentrations from stronger winds.

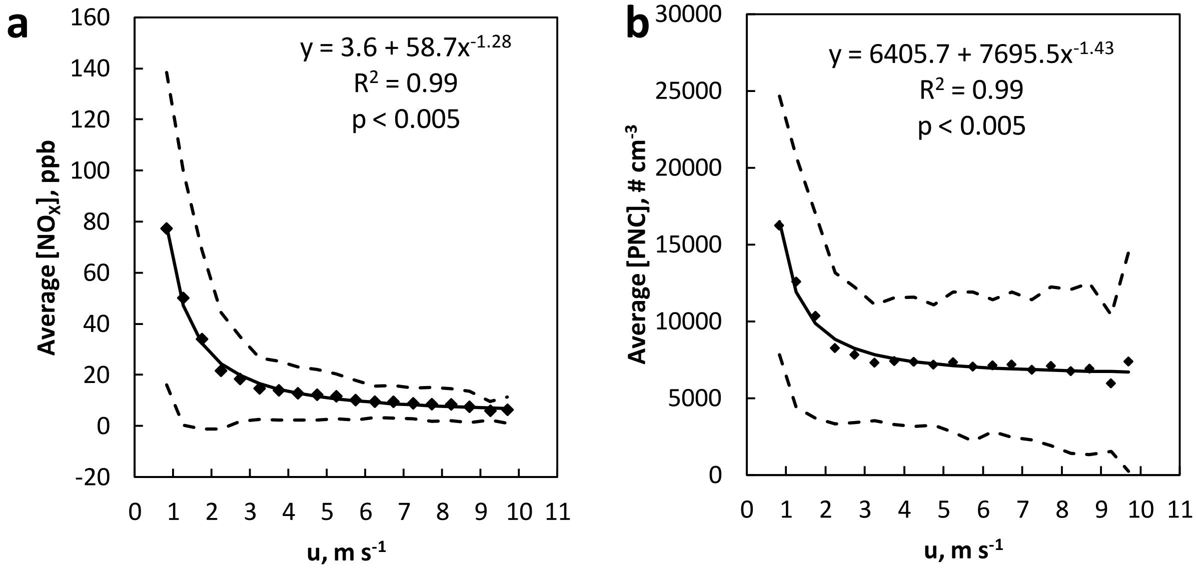

Figure 3. Relationships between wind speed (*u*) and the concentration of (a) NO_x and (b) PNC. Each point in the relationships is an average for a wind speed interval with steps 0 – 0.5, 0.5 – 1 m s^{-1} and so on. The dotted lines show the standard deviation for each interval. The relationships are based on the least squares method and statistical significance is based on the F-test.

For PM_{10} a slight increase in average levels was observed for increasing winds although not reaching as high as during low wind speeds (**Paper III, Figure 2d**). At high winds PM deposited on dry surfaces may re-suspend which could partly explain the slight increase in levels. The accumulation of ground level air pollution emissions is also strongly related to the atmospheric stability of the PBL. The degree of atmospheric stability was represented by the vertical temperature gradient and also showed strong relationships with the variation in levels of air pollutants (**Paper III, Figure 3**). At well mixed situations ($\Delta T < 0°C$), when the PBL reaches large height, pollutants are vertically well dispersed and pollution levels near the ground tend to decrease. Stable conditions were defined as a positive vertical temperature gradient ($\Delta T > 0°C$) and highest average levels were found for all tested pollutants (NO_x, NO_2, PNC and PM_{10}), indicating that the lack of vertical air mixing traps air pollutants near the ground.

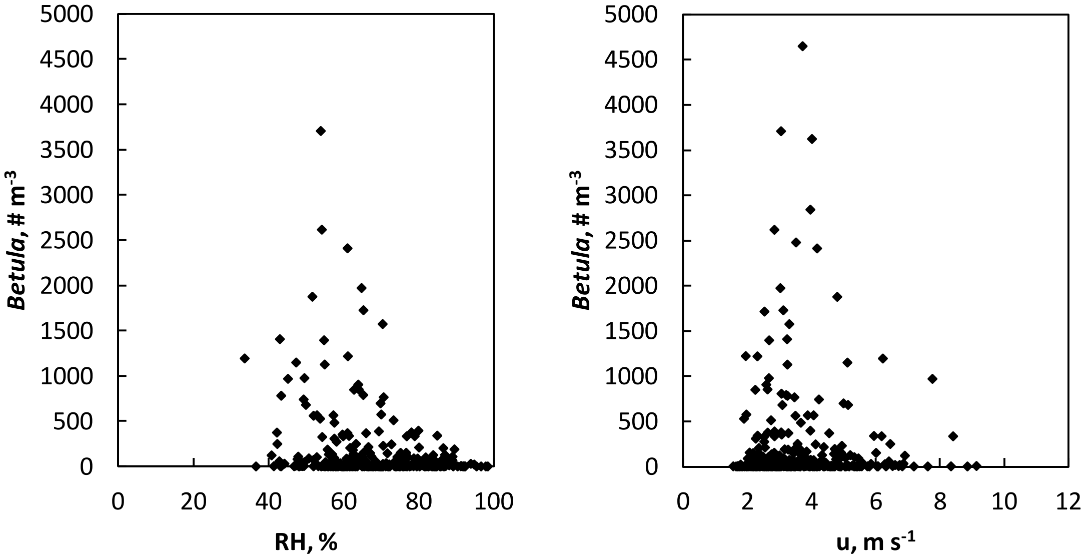

Figure 4. Daily birch (*Betula*) pollen counts (Gothenburg) in relation to (a) the relative humidity and (b) wind speed during pollen seasons 2006-2012. RH = relative humidity, u = wind speed.

During spring months when the birch tree starts to flower dry conditions are important for the release and dispersal of pollen. Anthers bursts in dry conditions and the wind helps expel it further into the air. In Figure 4, high levels of daily birch pollen counts showed a strong dependence on dry conditions. A large fraction of the high levels were observed mainly at low relative humidity (RH < 80%) and low to moderate wind speed ($2 < u < 4$ m s^{-1}).

4.2 Atmospheric circulation influences local meteorological conditions

4.2.1 NAOI and LWTs influence meteorology in winter

The typical local meteorological character of circulation patterns were categorised using synoptic scale circulation types (LWTs) and the large scale circulation represented by the NAOI. During winter (December, January and February) the monthly NAOI was directly characterised with the prevailing meteorological conditions in Gothenburg for 1997-2006 (**Paper I**). It was found that the positive phase of the NAO (NAOI > 0) more often was associated with low atmospheric pressure (Figure 5a), mild temperatures and surface winds more often from west and south. The negative phase (NAOI < 0) was associated with high atmospheric pressure, cold temperatures and wind directions more often from north, east and south. Simplified, the two NAOI phases can be associated with two types of weather regimes, mild, low pressure weather or cold high-pressure weather. Furthermore, LWTs calculated for Gothenburg, were also directly tested in relation to the NAOI for 2001-2010 (**Paper II**). Westerly LWTs (SW, W and NW) were generally very common under the positive NAOI, confirming its strong association with the westerly wind flow over the Atlantic. Under negative NAOI the occurrence of these types were significantly reduced while LWT C increased (Figure 5b). In general, LWTs SW and W represented windy and mild low pressure weather, which was further enhanced during the positive NAOI.

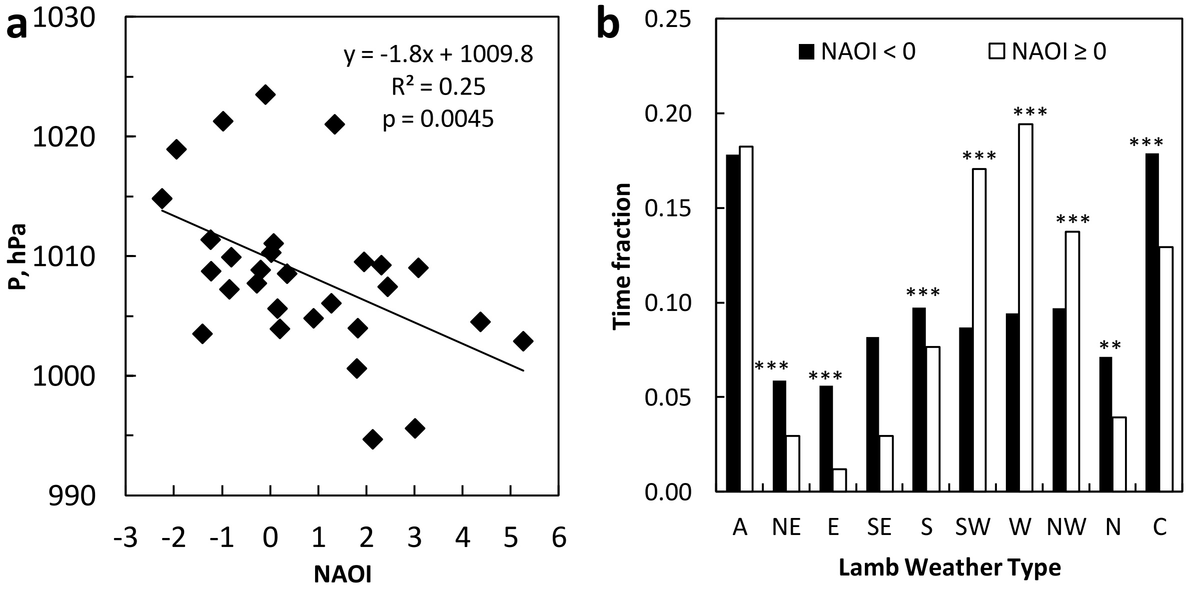

Figure 5. Linear regression between monthly means in NAOI and atmospheric pressure (P) during winter 1997-2006 (a) and the frequency of LWTs during positive and negative NAOI during winter 2001-2010 (b) in Gothenburg.

During negative NAOI, all LWTs tended to be colder and calmer than during positive NAOI **(Paper II, Table 1)** and the occurrence of northerly to southerly LWTs (NE, E, SE, S and N) increased together with LWT C. Thus, a negative NAOI is associated with a lower occurrence of SW and W thus also a lower occurrence of wet and windy weather transported from the Atlantic. LWT A was relatively common in both phases of the NAO but colder during negative NAOI. Overall the NAO in winter can be linked to the weather conditions in Gothenburg, through its direct association with the local meteorological situation and through the representation of circulation types (LWTs). Cold weather was associated with LWTs A, NE, E, SE and N while calm weather conditions were associated with LWTs A, N, NW and C.

4.2.2 LWT influence on meteorology on a yearly basis and during the spring

The meteorological character of LWTs was further investigated on a yearly basis **(Paper III)**. Figure 6a shows the meteorological character of LWTs from April 2007 – April 2008. Calm wind speeds ($u < 2$ m s^{-1}) and stable weather ($\Delta T > 0.5$°C) was most often associated with anticyclonic conditions and air masses transported from polar and easterly continental areas i.e. LWTs A, NW, N, NE and E. Due to the variation in temperature throughout seasons the meteorological pattern will show some variability. An example was LWT NW which tended to be more calm relative to other LWTs during winter (**Paper II, Table 1 and Figure 3**) than during spring and year (Figure 6b). Windy and wet weather was strongly coupled to LWTs SW, W and C on both yearly and seasonal basis. During spring LWT S was associated with wet weather despite its dry character, signified by its association with a higher than average VPD (> 0.55 kPa) as can be seen in Figure 6b.

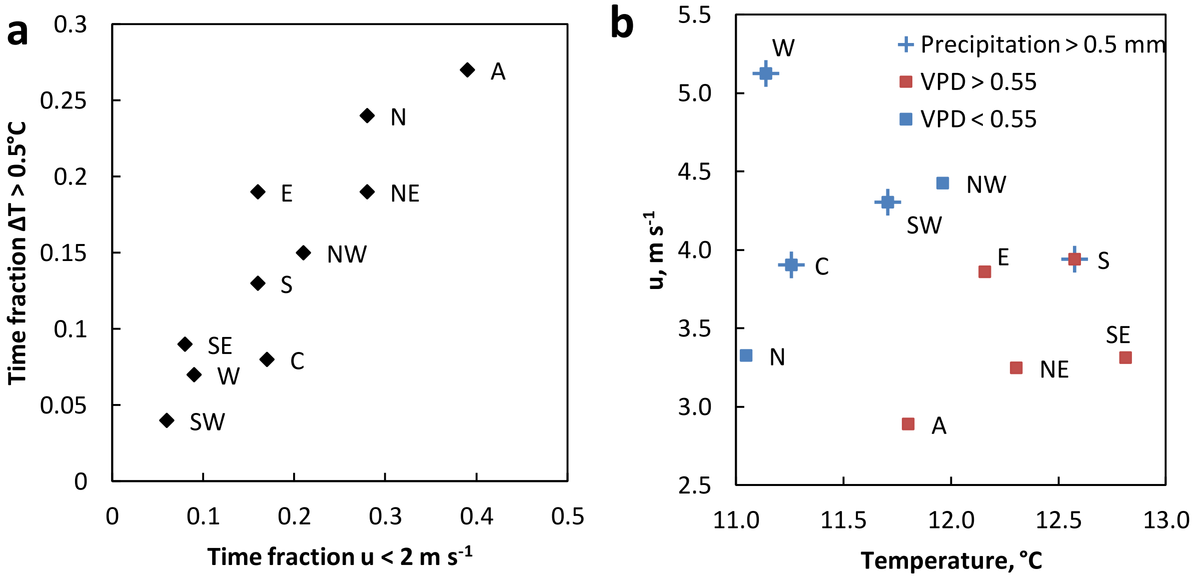

Figure 6. Meteorological characterisation of LWTs (a) based on the relative frequency of low wind speeds (< 2 m s^{-1}) and stable conditions ($\Delta T > 0.5$°C) April 2007 – April 2008 and (b) during birch pollen season (*Betula* > 0) for 2006-2012.

During the spring season when the birch tree starts flowering, warm, calm and dry conditions are important for the pollen to mature and disperse. In **Paper IV** the meteorological character of LWTs were based on spring months (March-May) 2006 – 2012. In Figure 6b a clear contrasts based on average meteorological conditions is distinguishable between LWTs. Warm, calm and dry (VPD > 0.5 kPa) conditions were represented more often by A, NE and SE. LWT E was generally warm and dry but windier than average and LWT N was relatively calm but wetter and colder than average. Precipitation was larger than the average for LWTs W, SW, C and S, linking them strongly to wet deposition. LWTs were also analysed for the city of Malmö, here the meteorological pattern was somewhat different in comparison to Gothenburg. There were also smaller differences in meteorological averages between LWTs in Malmö, i.e. less variation in their meteorological character. This can be seen in **Paper IV, Figure 1**.

4.3 High exposure situations are influenced by the atmospheric circulation

4.3.1 NO_2 levels in relation to NAOI and LWT during winter

Since the two phases of the NAOI and all LWTs show distinct differences in their association with windy or calm weather conditions, it can be expected that air pollution levels also show a distinct variation in relation to these weather patterns. Focusing only on winter and NO_2 the direct influence from the NAOI was very clear **(Paper I)**. Figure 78 shows the diurnal variation of high NO_2 levels represented by the time fraction of exceedances of NO_2 AQS during negative (NAOI < 0) or positive (NAOI > 0) NAOI. Exposure to high levels is clearly more probable during morning rush hours (7-10 am) especially when the NAOI is weak.

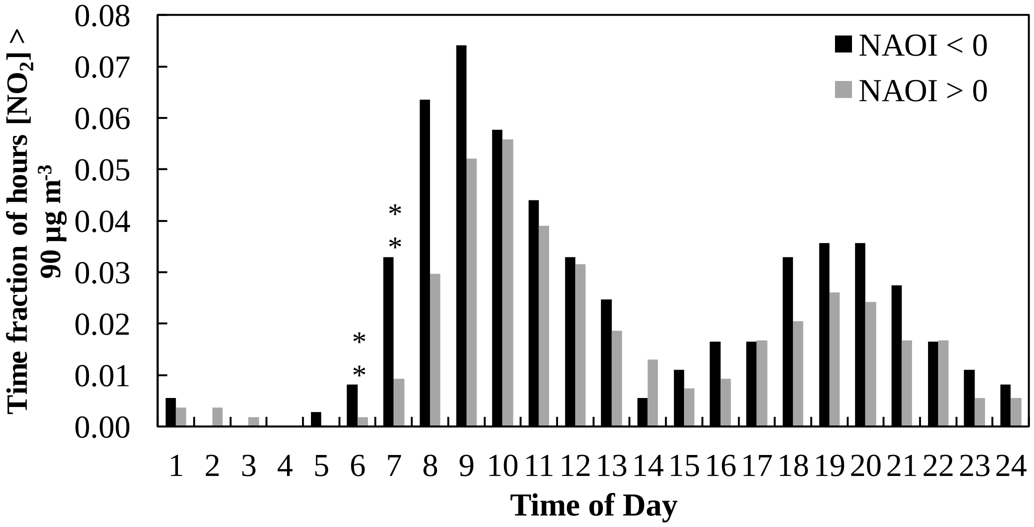

Figure 7. Diurnal variation in NO_2 exceeding 90 AQS during negative (black bars) and positive (grey bars) NAOI, during winter months (January, February and December) in the period 1997-2006.

The variation of high NO levels showed a similar but clearer diurnal pattern with more hours showing larger significant differences between positive and negative NAOI (**Paper I, Figure 5b)**. The bi-modal pattern of diurnal air pollution levels is commonly observed in urban environments where traffic is a dominant emission source. **Paper I** also revealed that more extreme negative NAOI values (NAOI < -2) resulted in even larger levels of NO_2 and NO than if all negative NAOI values were considered. This indicates that a very weak NAO can have a more severe effect on air pollution levels. In **Paper II**, the NAOI association with the occurrence of LWTs and their respective and combined effect on NO_2 levels were analysed. Figure 8 shows that LWTs occurring during negative NAOI were associated with larger fractions of exceedances of the hourly NO_2 AQS (NO_2 > 90 µg m^{-3}) at the urban ground level site than for LWTs occurring during positive NAOI. Furthermore exceedances of NO_2 AQS showed a strong pattern in relation to frequency of low wind speeds specific for LWTs **(Paper II, Figure 3)**. The pattern was very similar for both the roof top and ground level site both located in the city centre of Gothenburg but with somewhat different surrounding landscapes. Exceedances were generally more common for LWTs associated with high fraction of light winds ($u < 1.5$ m s^{-1}) and LWT N and NW showed the highest probability for exceedances of NO_2 AQS. It was shown that LWTs and NAOI had both independent and common effects on NO_2 levels in Gothenburg.

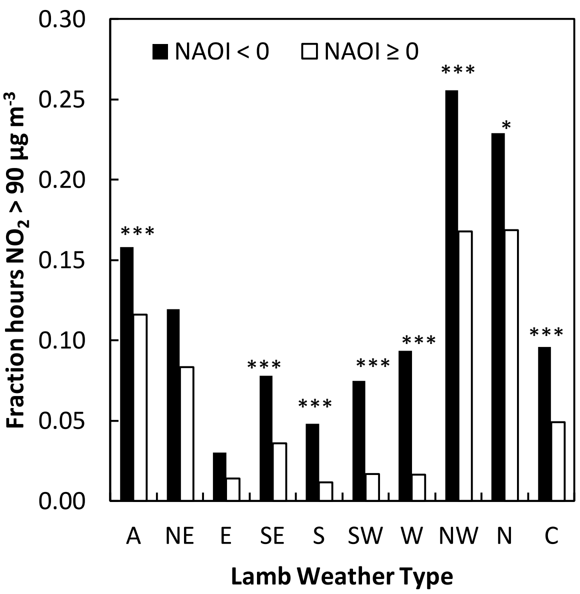

Figure 8. Fraction of exceedances of NO_2 > 90 during different LWTs at an urban kerbside site in Gothenburg during winter months January, February and December for 2001-2010.

4.3.2 NO_x - an efficient proxy for PNC (UFP)

UFPs have received increased focus for their potential large contribution to air pollution effects on human health. However, there are no direct AQS regulating the levels of UFPs, hence there is a lack of extensive measurements and data. There are AQS for PM_{10} but this measure is a poor approximation of UFPs, since UFPs are dominated by particle number rather than particle mass. Hence, UFPs contribute little to the mass based PM_{10}. PNC measurements can serve as a UFP substitute since PNC is dominated mostly by UFP. In **Paper III** an air pollutant proxy for PNC in the urban environment was investigated. Concentrations of PM_{10} were generally poorly related to PNC, the daily PM_{10} had a weak ($R^2=0.17$) correlation with PNC and can be seen in Figure 9a. For daily averages of PNC-NO_x good positive relationships were found, especially during days with temperature inversions ($R^2=0.73$), similar patterns were observed for PNC-NO_2, but with somewhat weaker correlation coefficients.

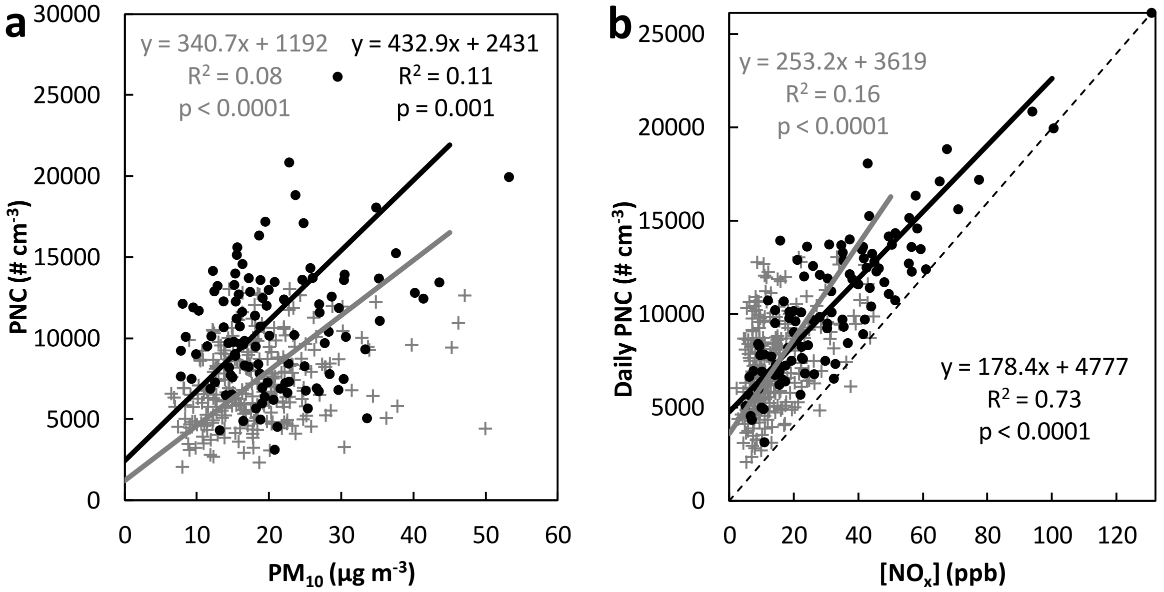

Figure 9. Relationships between daily averages of PNC and (a) PM_{10} and (b) NO_x in Gothenburg from April 2007-May 2008. Grey plus symbols signify observations occurring during well mixed situations (ΔT < 0°C) and black dots signify observations occurring during temperature inversions (ΔT > 0°C).

In other words, situations associated with high levels of NO_x and/or NO_2 can be expected to also have high PNC. When including the effect of meteorology the proxy relationship was especially strong during low wind speeds ($R^2=0.82$), high vertical temperature gradients ($R^2=0.85$) as can be seen in **Paper III, Figure 4a and d**. This again verifies their similar response to calm and stable conditions. Additionally, when aggregating hourly observations into averages for ten LWTs the PNC-NOx relationship remained very strong (Figure 10a). When only considering observations with a prevailing positive temperature gradient the relationship was improved further ($R^2=0.93$) as can be seen in Figure 10b. It is obvious that LWTs A, N, NW and NE are associated with the higher average levels of both NO_x and PNC. These LWTs were also associated more often with low wind speeds, important for high levels of NO_x and PNC.

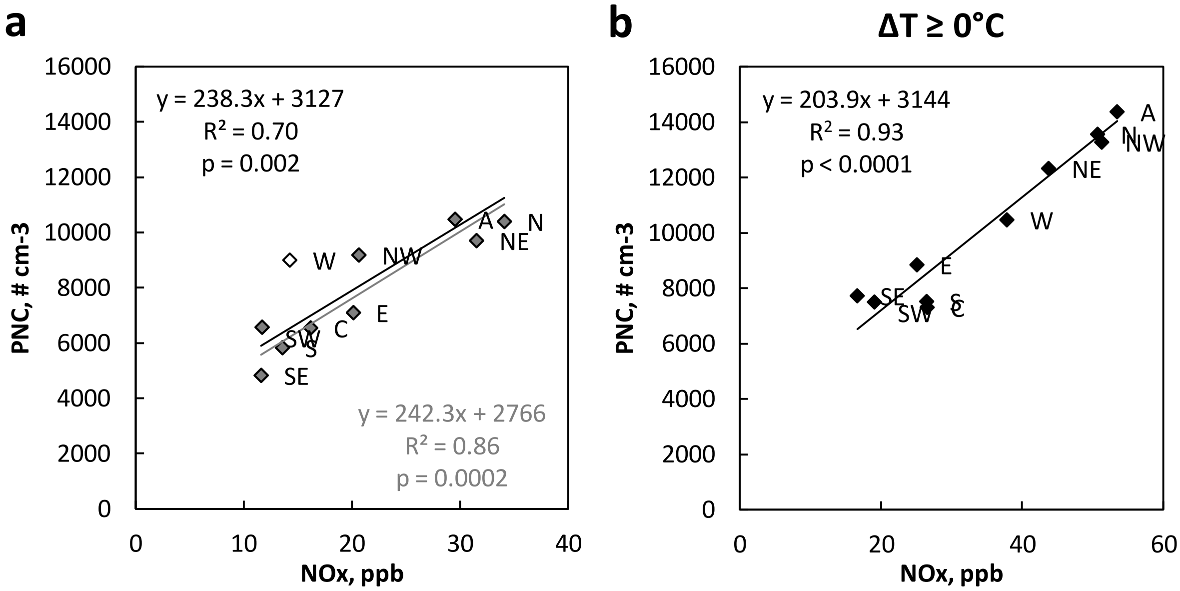

Figure 10. Relationship between LWT averaged concentrations of PNC and NO_x in Gothenburg from April 2007-May 2008. The grey regression line in the left panel excludes LWT W from the relationship. In the right panel only observations with inversion days were included (ΔT > 0°C).

High NO_x and NO_2 are promoted by the same meteorological conditions i.e. low wind speed and a positive (inverted) vertical temperature gradient as was demonstrated in **Paper III**. PNC showed very similar response patterns to these meteorological factors, while PM_{10} showed a different pattern. An interesting observation was also that PNC was poorly related to NO_x during high wind speeds ($u > 5$ m s^{-1}) and the relationship differed in comparison with the two relationships based on lower wind speed categories (**Paper III, Figure 4d**). PNC was moderately high during high wind speeds while NO_x were relatively low indicating a potential separate PNC regime. A deviation was also observed for LWT W from the regression between averages of NO_x and PNC for LWTs as can be seen in Figure 10a. LWT W was associated with a large frequency of high wind speeds (**Paper III, Table 1**). When excluding LWT W from the relationship (grey regression in Figure 10a) it was improved ($R^2 = 0.86$). Furthermore, when only considering observations with stable conditions in the respective LWTs the PNC-NO_x relationship was further improved ($R^2 = 0.93$) as can be seen in Figure 10b.

4.3.3 Co-variation between air pollutants and birch pollen during LWTs

In spring, pollen levels increase in the ambient air and cause health problems for people suffering from pollen allergy. In the urban environment exposure to both high air pollution levels and pollen take place which can result in enhanced effects on human health. In **Paper IV** the co-variation between high levels of birch pollen count together with NO_2, O_3 and PM_{10} was investigated during the influence of LWTs. Figure 11 shows an example of the average birch pollen levels for LWTs. A clear pattern was apparent with the highest birch pollen levels observed during LWTs characterised by warm, dry and moderately calm conditions, especially during LWTs E and SE.

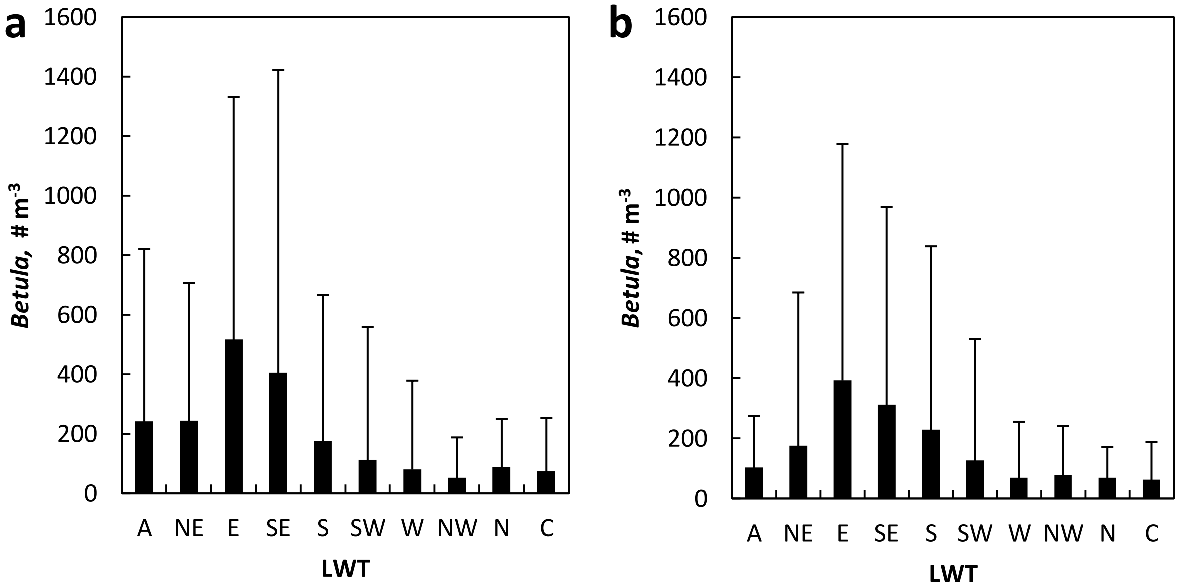

Figure 11. Average daily birch (*Betula*) pollen counts for different LWTs in Gotehnburg (a) and Malmö (b) during the birch pollen seasons (*Betula* > 0 # m^{-3}) between 2006-2012.

These LWTs exhibit low to moderate winds simultaneously with dry conditions. Both characteristics promote high pollen concentration but also high air pollution. The lowest average birch pollen levels were found in wet and windy LWTs SW, W and C together with NW and N. When considering the frequency of high daily birch pollen counts (*Betula* > 100 grains m^{-3}) it was obvious that high levels of air pollutants together with birch pollen co-varied on an LWT basis. Figure 12 shows strong relationships between the time fractions of high daily birch pollen counts and high daily maxima of NO_{2max} (R^2= 0.69) and PM_{10max} (R^2= 0.72) in Gothenburg **(Paper IV).** In other words, certain LWTs are associated with a higher risk of exposure to both high pollen levels and high air pollution levels. In Malmö there was no significant co-variation observed between air pollution and birch pollen (**Paper IV, Figure 7 d-f**). Furthermore, it was shown that for 70-90% of the ten LWTs, sales of OTC antihistamines (used as a proxy for health symptoms) was higher for situations with simultaneous occurrence of high birch pollen and high air pollution (NO_2, PM_{10} and O_3) levels compared to high pollen and low air pollution levels, in both cities (**Paper IV, Figure 8d-f and 9d-f**). This indicates a potential enhanced risk for negative health effects.

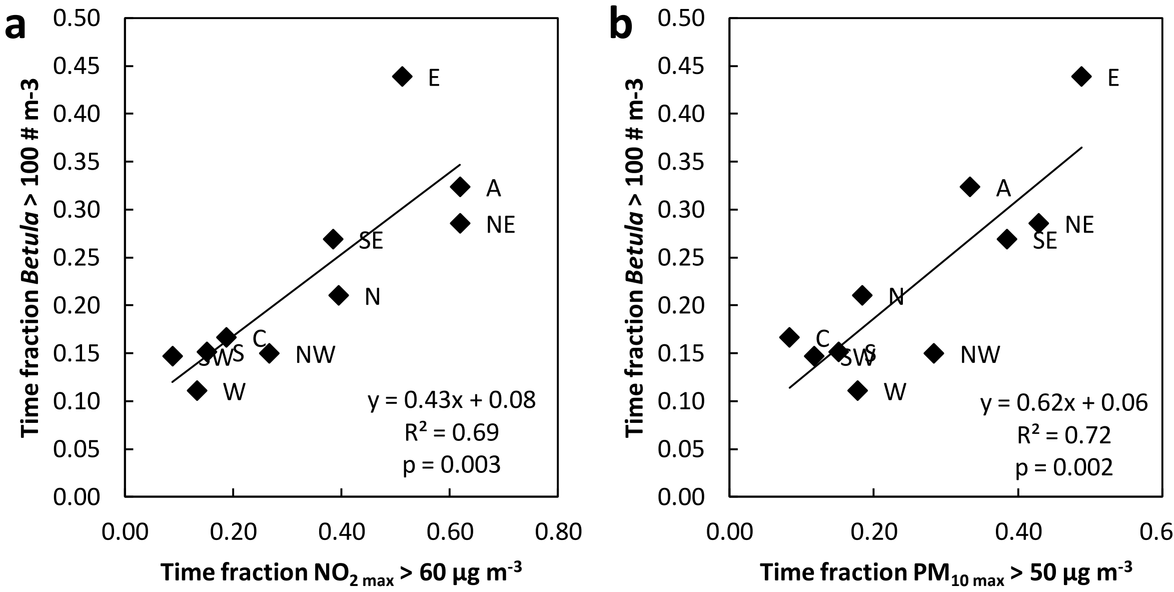

Figure 12. The relationship between fractions of exceedances for daily birch (*Betula*) pollen counts and daily maxima of NO_{2max} (a) and PM_{10max} (b) in Gothenburg during birch pollen seasons 2006-2012.

4.4 Anomalies in air pollution levels link to anomalies in weather

Air pollution levels and the compliance with legislated AQSs are evaluated on a yearly basis; therefore an important application of the LWT method is to test whether it can be used to evaluate the year-to-year variation in air pollution levels. A year with a high frequency of wet and stormy weather can be expected to result in lower levels of air pollution without any emission reductions taking place. In **Paper V** the year-to-year variation in air pollution levels was investigated as a function of the year-to-year frequency distribution of LWTs. The variation was expressed as anomalies i.e. negative or positive deviations from the normal average valid for 1997-2010. Figure 13a shows, as an example, the anomalies of annual average [NO_2] as a function of anomalies of the annual LWT combination e.g. the sum of the relative frequency distribution of each LWT for each year. It was obvious that a high anomaly in average [NO_2] was explained by high anomalies in LWT, meaning that certain LWTs, known to promote high [NO_2] were relatively common that particular year.

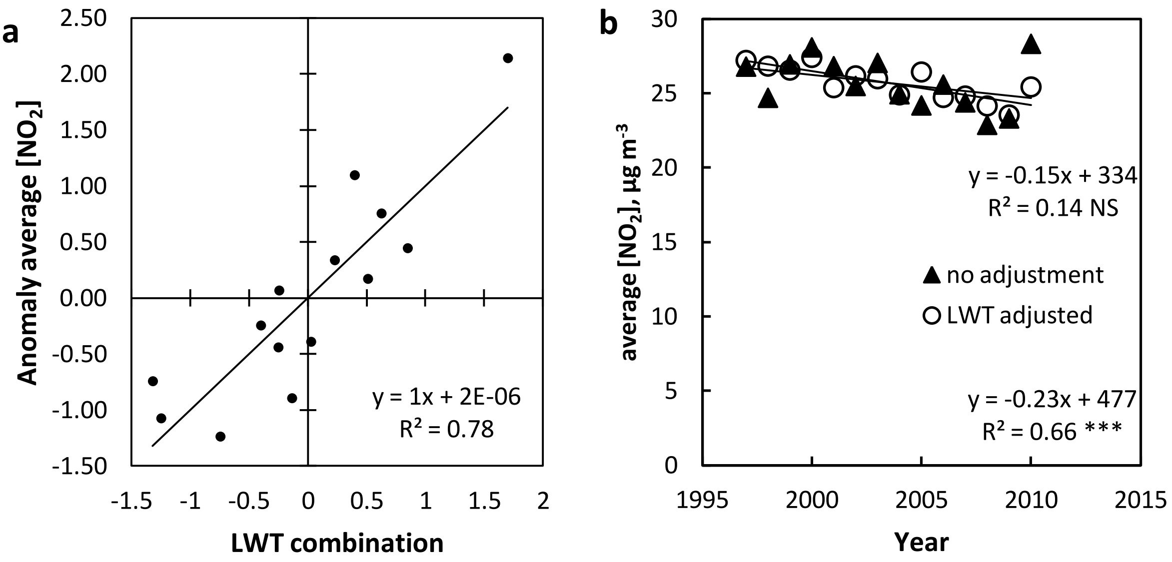

Figure 13. Observed annual anomaly vs the anomaly resulting from the optimized LWT combination for average [NO_2] (a). Temporal trend for observed and LWT adjusted average [NO_2]. 1997-2010, Gothenburg (b).

Figure 13b shows a non-significant negative trend in NO_2 from 1997-2010 (no adjustment). When adjusting the yearly averages to the variation in weather for each year, the trend was strongly statistically significant and shows that a large part of the negative trend is explained by the weather conditions. The deviating unadjusted observation for year 2010 was clearly reduced after LWT adjustment. That year was a particular cold year due to the extremely cold and long winter with several high NO_2 pollution episodes. Other atmospheric compounds were tested (PM_{10}, CO, O_3, deposition of SO_4^{2-}, NO_3^- and NH_4^+) and the LWT system was successful in explaining a large fraction of the inter-annual variability. Those results are outlined in more detail in **Paper V**.

5. Discussion

5.1 General remarks

On the whole, this thesis has shown that local scale meteorology together with synoptic and large scale circulation can be linked to urban air pollution levels of NO_2 (**Paper I - IV**), NO_x**,** PNC**, (Paper III),** PM_{10} **(Paper III, IV)** and birch pollen counts (**Paper IV**). The variation in wind speed and vertical temperature differences had strong relationships with the urban air pollution levels. Limited ventilation represented by low wind speeds and positive vertical temperature gradients were associated with high levels of NO_2, NO_x, PNC and PM_{10} (**Paper III**). A number of studies are in line with these observations where low wind speeds and inversions were associated with high levels of different pollutants (Janhäll et al., 2006; Jones et al., 2010). Furthermore, these conditions can be associated with the atmospheric circulation both on a large scale (NAOI) and on a synoptic scale (LWT). Certain air masses associated with specific LWTs (**Paper II, III, IV**) and/or a negative NAO (**Paper I, II**) were more frequently associated with limited ventilation conditions and hence higher levels of air pollutants. During the winter calm, dry and stable weather situations were more often represented by the negative NAOI (**Paper I, II**) and LWTs A, NW and N (**Paper II).** Studies from Belgium, Australia, UK and Portugal have also showed strong links between air quality and synoptic weather using different weather type classifications (Demuzere et al., 2009; Pearce et al., 2011; Pope et al., 2014; Russo et al., 2014). Furthermore, the strength of the proxy relationship between PNC-NO_x (**Paper III)** was influenced by meteorological variables and LWTs i.e. the strength of this relationship is dependent on weather. This is an important finding in terms of health effects related to PNC (UFP) which lacks direct regulation and is poorly related to PM_{10} (as shown in **Paper III**).

Deviations from the typical meteorological pattern.

The typical meteorological character of LWTs was not fully consistent. There are periods when deviations from the characteristic pattern occur both within the same season and in between seasons. This was observed for westerly to south westerly air masses represented by LWT W and SW. These had periods of calm winds indicated by their (low) time fraction of low wind speeds (**Paper II, III**). From an air quality risk assessment perspective these situations are important to note even though they were relatively uncommon.

Seasonal patterns in meteorological character and air pollution

On a yearly basis LWTs A, NE and N were the top three LWTs more often associated with calm conditions and also high air pollution levels (NO_x and PNC, **Paper III**). Also LWT NW was associated with calm weather on a yearly basis, but to a larger extent during the winter together with LWT A and N (**Paper II**). This explains why LWT NW was associated with lower NO_x levels on a yearly basis in comparison with LWTs A, NE and N. Furthermore, this highlights the fact that each LWT may exhibit some seasonal differences in their meteorological character thus also on processes governing high or low air pollution levels. Additionally, air pollution levels also have a seasonal cycle, partly determined by meteorology but also due to changes in emission patterns throughout the year. NO_2 peaks in winter, PM_{10} and birch pollen in the spring and ozone during spring and summer and from a scientific perspective air pollution levels should be evaluated in relation to weather conditions also on a seasonal level to account for seasonal variation in both air pollution and weather conditions associated with specific LWTs. Despite seasonal variation, the inter-annual variation in average air pollution levels showed strikingly strong relationships with anomalies in LWT combination (**Paper V)**, further demonstrating their usefulness in assessing the weather effect on urban air quality.

Local aspects may be of importance for concentrations

Directly north east of the urban roof top site in Gothenburg, the central bus terminal is located which could potentially have a positive influence on NO_2 levels. In winter high NO_2 levels were observed in LWTs NW, N and A (**Paper II**), the same pattern was observed for the street level site with a different surrounding landscape, without a specific nearby source directly in north-east. In spite of this the pattern remained similar with high NO_2 levels in A, NW and N i.e. the local source seems to be of minor importance, as a different pattern would be expected for the roof top site if the local emission source was of large importance. In spring however, LWT NE and E were associated with relatively large time fractions of high NO_2 levels while LWT NW was not (**Paper IV**). This is partly explained by the lower average wind speeds associated with LWT NE during spring in comparison with NW. LWT E however, was associated with more windy conditions together with surface wind directions often from the north-east. North-easterly surface winds were coincidently more often associated with high hourly NO_2 levels, thus a contribution from the local bus station cannot be completely ruled out, even if both urban sites have many traffic air pollution sources in all directions.

The deviating PNC regime less associated with NO_x during ventilated conditions and LWT W, found in **Paper III,** could be a potential contribution from marine salt particles from the sea located directly west of Gothenburg. Significant sea salt transport across southern Sweden was demonstrated by Gustafsson and Franzén (2000). Different PM sources are present and are likely to influence the PNC-NO_x relationship.

Long range transport or local source contributions

Directional LWTs are undoubtedly associated with some degree of transported pollutants from other regions. However, the atmospheric life time of some compounds are quite short and therefore not subjected to substantial transport over large distances. NO_x and NO_2 have a life time of 1-2 days in the atmosphere and are considered to be a local pollutant (Finlayson and Pitts, 2000). This was demonstrated by the large differences in NO_2 concentrations between the ground level and roof top site in **Paper II**. The lifetimes of pollen and PM_{10} span from tens of hours to a couple of days and are thus more subjected to atmospheric transport over larger distances. Air masses travelling from continental Europe are expected to contain the most polluted air due to the large number of urban areas and other sources in this region. However air mass flow from these regions (S, SW and partly SE) were mainly associated with small fractions of high levels of NO_2 (**Paper I-IV**) and PM_{10} (**Paper III, IV**) and low average levels of PNC (**Paper III**). These types were often associated with strong winds ($u > 5 \text{ m s}^{-1}$), conditions known to both aid transport but also ventilates the air of locally emitted pollutants (**Paper III**) and birch pollen. The fact that low levels of air pollutants were observed for LWTs S, SW and SE indicate that the meteorological properties of these LWTs are more significant for ventilating the air than the potential contribution to high pollution levels from long-range transport.

For ozone, episodes of high levels in Sweden are known to often be of non-local origin and the high levels of O_3 observed in southerly LWTs (**Paper IV**) are well in line with previous studies showing the importance of ozone transport from other regions (Tang et al,. 2009, Solberg et al., 2005). For background air pollution levels long range transport may have a more significant impact, Johannesson et al. (2007) showed that background levels of $PM_{2.5}$ in Gothenburg increased during days when the air mass over the region had been transported from continental Europe, similarly PM_{10} levels in **Paper III** may have been influenced from transport but relatively low average levels were found in southerly LWTs. Pollen transport from regional to larger distances have also been evident (Skjøth et al., 2007, Ranta et al., 2006, Sikoparija et al., 2013) and are potentially of importance for high levels. Dry and moderately calm LWTs E and SE during spring were associated with high birch pollen levels in both Gothenburg and Malmö which are assumed to be an effect of local, regional and long-range transport. The relative importance of each scale is difficult to assess, especially during the peak of the local pollen season. Trajectory analysis can contribute to identify potential source regions but again the meteorology controlling dispersion at the source regions and during transport need consideration to estimate the potential source contributions. In any case, the risk of high pollen levels without considering specific source contributions can be assessed using LWTs.

Climate change perspective

The strong connection between atmospheric circulation and air pollution shown in this thesis poses implications in a future climate perspective. Jacob and Winner (2009) found a weakening of the general circulation and that the frequency of episodes of stagnation will increase over northern mid-latitude continents. Stephenson et al. (2006) found that several general circulation models simulated a slightly increasing trend in the NAO. If this turns out to be correct, fewer wintertime episodes of AQS exceedance for NO_2 related to the NAO can be expected in the future if the emissions rates remain unchanged. However, if stagnant conditions increase and potentially increasing the frequency distribution of LWTs A, NW, N, NE and E throughout the winter or spring, high levels of air pollution and pollen can be expected more frequently. In any case, a shift in the climatic pattern has the potential to significantly affect air quality in Gothenburg and likely also other cities of e.g. North-West Europe, even if emissions remain constant.

The strong non-linearity of the relationships between air pollution concentrations and wind speed (**thesis Figure 3; Paper III**) shows that for local pollutants it is the potential change in frequency of very low wind speed ($u < 2\ m\ s^{-1}$) from climate change which is of importance. Thus, to evaluate the effect of climate change on local air pollution cannot be made by analysing the change in medium or high wind speeds, since air pollution concentrations are relatively constant over this range of wind speed, but increase strongly as wind speed reaches low values.

5.2 Concluding remarks

The strong association between air pollution, pollen and the weather represented by NAOI (**Paper I, II**) and LWTs (**Paper II-V**) found in this thesis emphasise their potential to be integrated into an objective risk assessment instrument for daily air pollution surveillance and warning systems. The LWT classification scheme provides a relatively simple method to characterise the weather thus grouping together many meteorological variables important for processes governing high or low levels.

Authorities responsible for air pollution management can benefit from the findings presented in **Paper V**. Yearly variation in air pollution levels are strongly connected to local emission changes and weather variability. Applying adjustments accounting for the weather dependency was shown to quantify a large fraction of its influence on air pollution levels. When the effects of annual weather variability were removed from the yearly air pollution, the trend relationships were in many cases greatly improved. This provides an effective method for air quality managers to further assess the role of other processes controlling air pollution levels such as emission changes.

From a health perspective the findings in **Paper III** could be of benefit when studying air pollution effects on health. Different pollutants are sometimes pre-supposedly or openly assumed to act as proxies for each other. This is partly based on the lack of measurements of all relevant air pollutants. There is no direct regulation of UFPs, thus not any extensive monitoring taking place, therefore limited empirical data. Many studies have shown that PNC can be used as a representative for UFP (Molnar et al., 2002; Charron et al., 2003; Kittleson et al., 2004). PNC is however poorly related to PM_{10}, thus health effects of PNC cannot be represented by PM_{10}. NO_2 has in many cases been used as a surrogate for traffic pollution (Gilbert et al., 2003) and as a surrogate for PNC (Seaton and Dennekamp., 2003; Eeftens et al., 2015) however the relationship between PNC-NO_2 found in **Paper III** was not as strong as the PNC-NO_x relationship, indicating that NO_x is an even better proxy for PNC, especially during calm and stable weather conditions. Integrating these findings in health studies may be of advantage.

6. Key Findings

Local meteorological variables strongly influenced processes governing high or low levels of air pollutants. Concentrations of NO_x, NO_2 and PNC had strong negative non-linear relationships with wind speed and positive linear relationships with temperature inversions. This emphasizes the strong dependence of high air pollution concentrations on dispersion.

Local meteorology was further represented by the synoptic atmospheric circulation using LWTs, each showing a distinctive meteorological character. During winter months LWTs with a westerly wind component (W and SW) and cyclonic vorticity (C) frequently represent mild, wet and windy conditions while LWTs A, NW and N frequently represent calm, cold and stagnant conditions, promoting high air pollution levels.

The frequency distribution of LWTs was influenced by the NAOI. LWTs A, SW, W and NW were the most common types during positive NAOI. The westerly LWTs were still common during negative NAOI, but had a lower frequency while LWTs NE, E, SE, S, N and C all increased in frequency in Gothenburg.

The variation in concentrations of NO and NO_2 and the number of exceedances of NO_2 AQS had a direct association with the variation in NAOI, without also considering local meteorological variables in Gothenburg. The NAOI had a negative relationship with NO_x and the negative NAOI was associated with large frequencies of exceedances of NO_2 AQS.

LWTs had a direct association with the variation in frequency of the number of exceedances of NO_2 AQS. The fraction of low wind speeds varied largely between LWTs and had a strong correlation with the time fraction of $NO_2 > 90\ \mu g\ m^{-3}$. LWTs A, NW and N were associated with the highest number of exceedances in winter.

LWTs and NAOI exerted both a combined and individual effect on NO_2 levels. When combined larger fractions of exceedances for most LWTs were associated with the negative NAOI.

Combining the influence of both LWTs and NAOI LWTs associated with frequent calm and cold weather conditions (A, NW and N) had higher frequencies of NO_2 AQS exceedances than other LWTs. Larger fractions of exceedances of NO_2 AQS is observed for the majority of LWTs occurring during negative NAOI than during positive NAOI.

PNC is a good proxy for UFP. However monitoring of PM_{10} is inadequate to represent UFP since the measure is a very poor proxy for PNC, at least in Gothenburg. There was a strong correlation between NO_x and PNC and the strength of this relationship was weather dependant, with the largest correlation coefficients during low wind speeds and temperature inversions. The association between NO_x and PNC remained strong for averages based on LWTs.

Birch pollen was shown to have a very strong weather dependency in both Gothenburg and Malmö with easterly and southerly LWTs having the highest levels.

OTC of sold antihistamines was related not only to birch pollen levels, but also to the combined occurrence of high birch pollen and high air pollution (NO_2, PM_{10} and ozone). These situations promoted OTC of sold antihistamines in almost all LWTs when pollen levels were high.

Annual anomalies for several air pollutants and deposition of sulphate, nitrate and ammonium could be explained to a large extent by accounting for the yearly variation of the mix of LWTs. Temporal trends of the pollutants became stronger when adjusting for the annual weather variability, which is useful e.g. when analysing trends following emission reductions.

7. Outlook

In this section a couple of examples are presented of how the findings of the present thesis could be used and further developed.

An important application of the weather effect on air pollution is to integrate it with mitigation strategies for improving urban air quality. When not complying with AQSs fines may be imposed on member states by the European Court. Coria et al. (2015) suggested temporally differentiated road pricing based on the rate with which pollution is dispersed to aid compliance with AQSs in Stockholm, Sweden. During rush hour and spring the probability for exceeding AQSs peaked and was strongly associated with the combination of traffic intensity and the ventilation capacity as represented by wind speed in Figure 14. The probability for exceeding the NO_2 90 AQS increased with increasing traffic intensity and with decreasing wind speeds. Thus, in days with low or very low wind speeds increased congestions charges in the city could be appropriate, since the contribution to high air pollution is much more likely compared to days with high wind speeds.

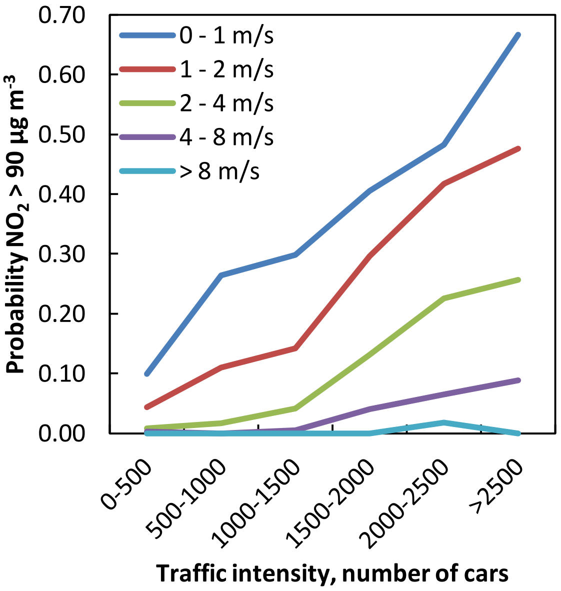

Figure 14. Relationship between traffic intensity and the probability to exceed the hourly NO_2 90 AQS during different wind speed categories, 2003-2007 in Stockholm, Sweden.

Introducing emission reduction strategies such as differentiated road pricing based on the weather effect may be one solution to lower air pollution levels. LWTs could be used in a similar approach to support policy tools for stimulating behavioural change and lower anthropogenic emissions especially during days when the risk of poor air quality is high. However, many societal aspects have to be considered when designing incentives for pollution reduction.

As it also was shown in this thesis, daily pollen counts were associated with specific LWTs. Forecasts including the weather effect represented by LWTs, as well as the possible additive impact of air pollutants to allergy symptoms, can aid risk assessment for high pollen levels, improve pollen forecasting and information to the public, and to identify the site-specific nature of adverse aerosol load in a certain region. The observed increase in antihistamine demand when incidents of threshold exceedances coincided for pollen and air pollutants could be quantified by the use of General Additive Models (GAMs). Figure 15 shows an example based on GAM analysis of how PM_{10} levels influenced the demand on antihistamines in both Gothenburg and Malmö.

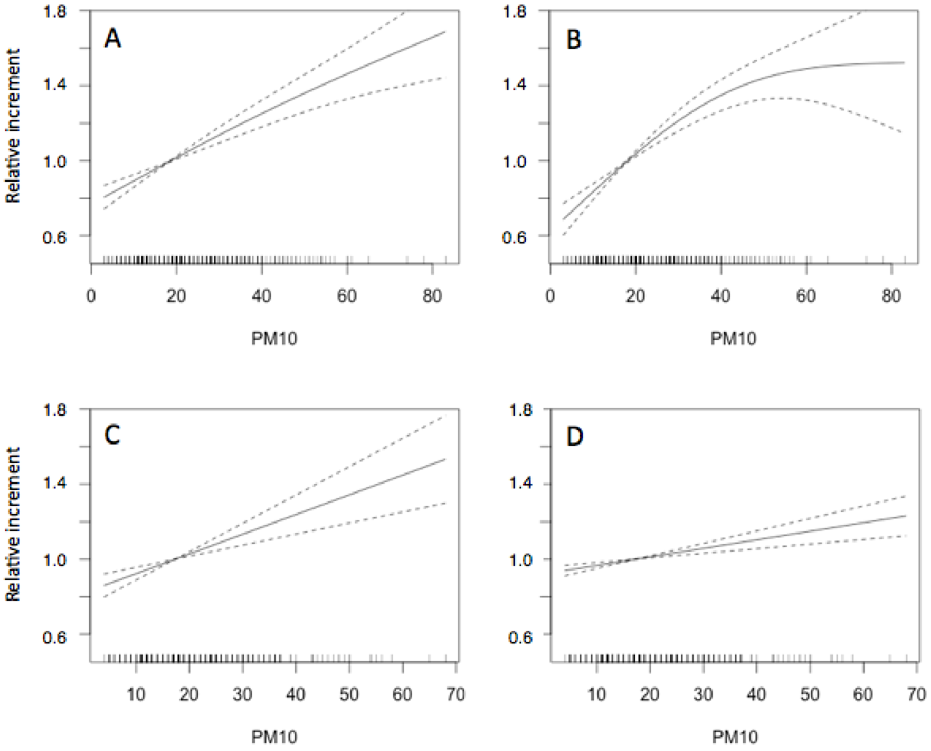

Figure 15. Estimated concentration-response curves (solid lines) and 95 % CIs (dashed lines), describing the effect of increment in ambient PM_{10} on the demand for antihistamines during the period April-August 2006-2012, in number of A. purchased defined daily doses (DDDs) over-the-counter in Gothenburg; B. prescribed DDDs in Gothenburg; C. purchased DDDs over-the-counter in Malmö and D. prescribed DDDs in Malmö. An increase with 10 $\mu g/m^3$ of PM_{10} will give 10% increase of OTC purchase and of prescriptions in Gothenburg, as well as of OTC purchase in Malmö ($p<0.0001$ for all), whereas the same increment of prescribed drugs in Malmö is 2% ($p= 0.00342$) . The relationship is calculated with general additive models (GAMs), which basically are general linear models (GLMs), in which it is allowed to incorporate smoothed functions. It is thus possible to remove" noise" from short-term variation in predictor variables, created by stochastic or unknown factors. Here, the smoothed function is a natural spline with four degrees of freedom.

These regression models allow for non-linear effects and for the control of confounding environmental variables. Furthermore, the inclusion of GAMs in a distributed lag model (Gasparrini et al. 2011) makes it possible to estimate the impact of pollutants, not only on the day when pharmaceuticals are dispensed, but also during the days or weeks previous to dispensation. Repeated exposure may add to the persistence of airway inflammation (e.g. Berhane et al. 2011). If such lagged effects are found, the impact of a certain LWT at a specific lag day could be evaluated. (Dahl, Grundström et al., work in progress).

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