Aerosol Measurements and Analysis

Categorization of Airborne Particles in an Urban Environment

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Samanfattning

Den största källan av skadliga luftburna partiklar i en urbanmiljö kommer från avgaser. Luftföroreningar har en stor påverkan på människors hälsa, uppskattad att orsaka 1.3 miljoner dödsfall per år världen över. Ungefär hälften av alla dödsfall orsakade av luftföroreningar kan kopplas till trafik. Men det finns flera andra källor som bidrar till vår förorenade luft från både naturligt och antropogent ursprung.

Syftet med denna kandidatuppsats är att bestämma storleksfördelningen på aerosolpartiklar, studera variationer i partikelstorlek och koncentration över tid och andra trender orsakade av meteorologiska förhållanden. Denna uppsats är en analys av data från en hälsostudie i Köpenhamn; vilket var ett sammarbete mellan hälsodepartementet i Köpenhamn och Ergonomi och Aerosol Teknologi vid Lunds Universitet. Vår analys av mätdata sträcker sig över perioden 20/12 2011-2/2 2012. Då denna period involverar nyårsafton kommer detta studeras närmare. Vi kommer använda tekniken från trajektorier för att detektera varifrån det blåser och partiklarnas ursprung.

Från ***SMPS*** datan kan vi se olika trender i storleksfördelningen och antal partiklar beroende på meteorologi och dagliga variationer. Nyårsafton visar en annan storleksfördelning än andra nätter; det är fler och större partiklar runt tolvslaget.

Antalsmedelvärdet skiljer sig inte så mycket mellan månaderna även om vi har två toppar med över 40,000 partiklar i december. I slutet av januari ökar volymen på partiklarna vilket korreleras till minskad temperatur och vindar från öst. När vinden byter riktning ser vi en ökning i partikelantal.

Högst antal partiklar hittas i storleksintervallet 10-100 nm, vilket stämmer väl överrens med teorin. Partiklar i detta storleksintervall är de mest skadliga för hälsan, då de är små och deponeras genom diffusionsmekanism djupt ner i lungan.

Det dagliga trafikmönstret är inte lika distinkt under rusningstrafik som vi trodde, istället ser vi en ökad koncentration dagtid. Detta kan bero på vägarbetet, den lokala topografin som innefattar ett öppet gaturum och de fyra sjöarna, vilket bidrar till en större uppblandning av partiklar.

När vi ser en ökad partikelmängd under natten beror detta på att vinden byter riktning. Vid östliga vindar ökar partikelstorleken vilket kan korreleras till långt transporterade partiklar som kan ha sitt ursprung från industrier i Tyskland, Polen och andra länder.

Abstract

The greatest source of hazardous airborne particles in an urban environment comes from traffic exhaust. Air pollution has a great impact on human health; estimated to cause 1.3 million deaths worldwide per year. About half of all mortality caused by air pollution can be linked to motorized traffic. There are several other sources that contribute to our polluted environment; from both natural and anthropogenic origin.

The aim of this bachelor thesis is to determine the size distribution of aerosol particles, study variations in particle size and concentration over time and study diurnal variation and other trends, caused by meteorological conditions. This thesis analysis is made on the data from an Exposure study in Copenhagen, which was collaboration between the Health Department in Copenhagen and Ergonomics- and Aerosol Technology, Lund University. Our analysis of the measurement data was made from 20/12/2011-2/2/2012. Since the period is over New Year's Eve, this specifically will be studied. The technique of trajectories is used to track the particles and try to determine where the air parcel comes from.

From the SMPS data we can see different trends in the size distribution and particle number depending on meteorology and diurnal variations. New Year's Eve shows a very different particle size distribution in comparison with other nights, the size of the particles is much larger this night and the number of particles shows a significant peak between 24.00-01.00.

We can see that the average particle number distribution does not differ so much between the three months, even though we have two peaks over 40,000 particles in December. In the end of January the volume increases and we can see a correlation with decreasing temperature and winds coming from East. We can observe larger amount of particles every time the wind changes direction.

The highest concentration of particles is in the range between 10-100 nm, which corresponds well with the theory. The particles in this range are the most hazardous to human health since they are small and deposit in the deep lung. This is favorable when doing human exposure studies.

The diurnal pattern in concentration due to traffic is not as distinct as we thought during rush hours; instead we have an increase in concentration during daytime. This can be due to road works and the local topography with the open street space and the four lakes, which contribute to a large mixing volume.

When we see an increase of particles during night time it can be correlated to change in wind directions. When the wind blows from the east we can see an increase in larger particles which is correlated to long range transport, likely explained by observations made of a lot of industries with strong emissions located in Poland, Germany and other locations where the wind has travelled over.

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1. Introduction

Air pollution is a very interesting topic, but most people do not know how hazardous these particles really are, and how many deadly outcomes they are responsible for. In this thesis focus is on small airborne particles called aerosols. The particle size in the aerosol spans over a wide range; from 1 to 100,000 nm. Despite their small size, aerosols can have a major impact on the climate and our health; if the aerosol concentration is high enough to be harmful it is classified as air pollution.

The greatest source of hazardous airborne particles in an urban environment comes from traffic exhaust. Urban outdoor air pollution is estimated to cause 1.3 million deaths worldwide per year *(WHO, 2011)* and about 320 000 deaths in 20 countries of Western Europe *(WHO, 2002).* About half of all mortality caused by air pollution can be linked to motorized traffic *(Künzli et al., 2000*). According to data in the IPCC 2001 Assessment report more than 90 percent of the aircraft emissions and approximately 99 percent of fossil fuel emissions are emitted into the atmosphere over the northern hemisphere *(IPCC, 2001).* The influence of long-range transported particles has been estimated to reduce the life-span of the Swedish population up to seven months, and cause about 3500 premature deaths per year *(Forsberg, 2005).* There are several other sources that contribute to our polluted environment from both natural and anthropogenic origin. How the particles are distributed over the globe depends on meteorology i.e. wind, temperature, altitude, and location of the particle sources. Differently sized particles have different properties and are therefore distributed in different layers in the atmosphere. Concentration, size and chemistry of the particles are believed to have the largest impact on human health.

The aim of this bachelor thesis is to determine the size distribution of aerosol particles (modes), study variations in particle size and concentration over time and study diurnal variation and other trends, caused by meteorological conditions. This thesis analysis is made on the data from an Exposure study in Copenhagen, which was collaboration between the Health Department in Copenhagen and Ergonomics- and Aerosol Technology, Lund University. This experimental study was set up close to a large road (Øster Søgade) in the central Copenhagen. The roadway was during the time of the measurements estimated to exhibit 50,000 vehicles per every 24 hours. The Human Exposure study aims to observe the accurate response- and inflammations caused by the outdoor air. In the study the Health Department stood for the human measurements on the exposure persons; people >60 years, with high BMI (>25, overweight), and-/or diabetes type II (non-medical treated) which is directly related to overweight. The team in Copenhagen made their analysis and tests after the persons spend their day in the exposure room. Lund University was responsible for the technical equipment, and the several instrument used to measure the particle concentrations, number size distributions, chemistry, particle effective density, mass, etcetera. The technique of trajectories is used to track the particles and try to determine where the air parcel comes from. Since the measurements are made over New Year's Eve, this specifically will be studied. During the millennium in Stockholm the amount of heavy metals in the atmosphere was five hundred times greater than normal, this due to fireworks *(Ny Teknik, 2000).*

Since this thesis is restricted to a period of time, the evaluation of the health study will not be ready before finalizing the report; and thus some limitation to its context had to be done. Therefore the health effects will not be covered. Focus is on the Scanning Mobility Particle Sizer (SMPS) instrument that measures particle number size distributions. In the laboratory set-up there were several other instruments that the scope of this thesis does not cover.

2. Background

2.1 Air pollution

Air pollution is defined as the presence of chemical compounds in the troposphere (i.e. the region below 11km) in large enough concentrations to harm organisms, climate or affect ecosystems. There are different sources of air pollutants; there are human sources (anthropogenic) and natural ones such as dust from e.g. windstorms, volcanos and pollutants from wildfires, even some kind of plants will release some volatile organic chemicals that can be harmful. The natural pollutants are distributed, removed and transported throughout our planet via different type of natural processes such as global cycles and gravity. Air pollutants have a large range of harmfulness, from irritating to lethal.

The human impact on airborne pollutants is largest in the urban countries, i.e. more people drive cars and industries are well established in these countries, see figure 1.

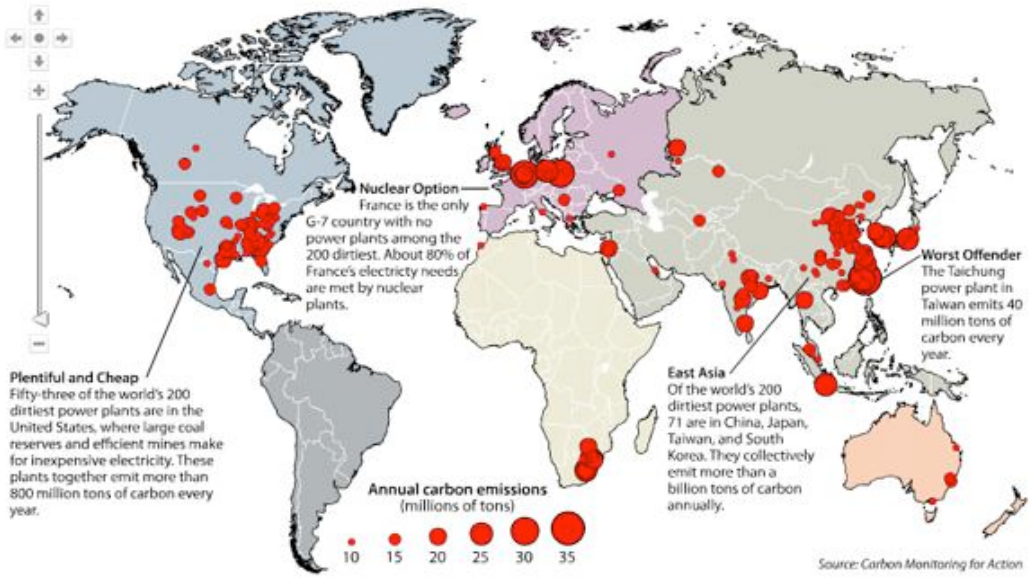


Figure 1. An illustration over the carbon emissions distributed over the globe. **(**http://thephoenixsun.com/wp-content/uploads/2009/1/1/Dirty-Coal-map.jpg).

Most of the gaseous pollutants comes from the combustion of fossil fuels in different ways; motor vehicles, power plants and industries. Pollutants can be divided into two sub-groups; primary- and secondary ones. Primary pollutants are those harmful compounds that are released directly into the atmosphere such as; CO, CO_2, NO, NO_2, SO_2. These compounds can react in the troposphere with either themselves or other components and then create a new form of pollutants called secondary pollutants such as; SO_3, HNO_3, H_2O_2, O_3 *(Miller, 2007, Ch.19).*

2.2 Aerosol particles

An aerosol is defined as a collection of solid or liquid particles suspended in a gas. The particle sizes in the aerosol spans over a wide range; from 1- 100,000 nm *(Baron et al., 2005, Ch.3),* see table 1. Despite their small size, aerosols can have a major impact on the climate and our health; if the aerosol concentration is high enough to be harmful it is classified as air pollution. This thesis focuses on aerosol particles which are normally found in our atmosphere, from both natural and anthropogenic sources, with main focus on the latter. Aerosols are always present in the earth's atmosphere and are part of the earth climate system; they are often very complex and consist of mixtures of particles with different size, shape and origin.

Aerosol particles are, as gases, divided into two categories; primary and secondary. Primary particles are emitted directly into the atmosphere e.g. sea salt, dust, and soot particles. A secondary aerosol has particles that are formed within the atmosphere for example organic particles that are formed from the oxidation of the Volatile Organic Compounds (VOC).

Aerosols can also be sorted into two different categories depending on their origin. Natural aerosols have the same origin as the natural gaseous air pollutants, and the same goes for the anthropogenic aerosols which are located in the lowest part of the troposphere over urban areas. Both natural and anthropogenic sources can be primary or secondary. In an urban environment the aerosols are a mixture of anthropogenic and natural sources, often dominated by the anthropogenic sources.

Type of particle	Size [nm]	Example
Aerosols	1-100,000	
Large particles	>10,000	
Cloud droplets	>20,000	
Ultrafine	1-30	
Fine particles	<1000	Soot
Nucleation mode	1-30	Fresh combustion particles
Aitken mode	30-50	Fresh combustion particles, Soot
Accumulation mode	50-1000	Biomass smoke, Marine organic,
Coarse mode	>1000	Dust, sea salt, pollen

Table 1. Table over the aerosol particles size.

Aerosol particles have varying properties such as particle size, concentration, chemistry, mixing status and physical shape. Aerosol particles are stable in the atmosphere at least for a few seconds up to a year or more, depending mainly on size. Large particles tend to have shorter lifetime than the smaller particles. For health and environmental effects the mass concentration and size are the most important properties to be studied and measured *(Hinds, 1999, Ch.1*), but other properties such as chemistry and surface area might also be important.

There are many uncertainties in the estimates of global emission of atmospheric aerosols, but it is known that overall the natural sources exceed the anthropogenic sources *(Hinds, 1999, Ch.14).* The natural sources are well distributed over the globe and often have its origin in large areas e.g. Sahara. Anthropogenic sources are fewer and more concentrated to smaller areas; where most population is located. Therefore in industrialized regions the anthropogenic sources contribute more to the particular mass than the natural sources. The two largest sources of natural aerosols is dust and sea salt; in the northern hemisphere dust contributes to 50-80 percent of the total mass of aerosols *(Gong et al., 2009, Ch.4).*

2.3 Formation, growth, transport and removal of aerosols

2.3.1 Condensation

The most important chemical atmospheric transformation of particles is condensation. It is responsible for the growth of atmospheric particles and for changing their chemical composition.

Condensation is defined as a process with more vapor molecules arriving at a particles surface than leaving the surface, resulting in a net growth of the particle *(Baron et al., 2005, Appendix A).* When a stable droplet of water is formed it can grow by condensation. The growth ratio depends on the particle size and gasphase saturation and it is controlled by the amount of vapor molecules arriving at the particle surface. When an aerosol exhibits a supersaturated condition (i.e. the relative humidity > 100 percent) the vapor condenses rapidly onto the particle population; which results in a change in the particle size distribution. The supersaturated environment is usually created by the gas-phase chemical reaction resulting in low pressure vapors or the cooling of vapors via atmospheric clouds *(Seinfeld et al., 1998).*

2.3.2 Nucleation

Homogeneous nucleation is the formation of a particle in a supersaturated environment without help from Condensation Nuclei (CN) or ions. The homogeneous nucleation process is rare in the atmosphere for water vapors, but can occur for other gases. If ions or nuclei are present the formation process is called nucleated condensation.

When supersaturated condition is fulfilled, a certain number of particles grow to produce fog or cloud droplets. This may partly occur also at sub-saturated conditions. Meteorologists call these particles condensation nuclei, or simply nuclei. Molecules in the supersaturated environment are more likely to collide with one another than in the saturated environment.

The greater the supersaturation is the larger number of molecules will collide and the concentration increases. Once the clusters reach a big enough diameter (d*~4 nm for water) it becomes stable and can grow by condensation, even if this diameter only lasts for a short time.

Heterogeneous nucleation/nucleated condensation are the process for particle formation or growth in the presence of nuclei or ions. This process relies on existing sub-micrometer particles, called condensation nuclei, to serve as sites for condensation *(Hinds, 2005, Ch.5).* Nucleated condensation can occur at much lower saturation ratios than homogeneous nucleation. If the droplets are stable the process can occur in environments that are unsaturated. Instead of homogeneous nucleation where the drops are formed by chance collisions of water vapor, molecule droplets are created by heterogeneous nucleation onto atmospheric aerosols *(Cotton et al., 2009, Ch. 2).* These droplets then grow by condensation and form cloud droplets at the super saturations achieved in natural clouds, these particles are then called Cloud Condensation Nuclei (CCN). Aerosol particles can be explained as the seed upon which water vapor condenses to form cloud droplets.

2.3.3 Evaporation

Evaporation is the reverse process to condensation which means that more molecules leave the surface of the particle than arrive. Evaporation is satisfied when the ambient partial pressure of vapor is less than the saturated vapor pressure; during evaporation a negative growth rate will occur which is a direct result of shrinkage due to evaporation.

2.3.4 Coagulation

When aerosols are formed they will undergo physical and chemical transformations in the atmosphere. The two most important physical transformations are collision and coagulation. Coagulation is an aerosol growth process which is a direct result of particle collision. During the process the number of particles decreases and the average particle size increases. This means that the net result is a continuous decrease in number concentration. The speed of the coagulation process is size dependent since the diffusion velocity is high for small particles and therefore small particles have short lifetime. The coagulation processes is the most important process between aerosol particles *(Hinds, 1999, Ch.13).*

2.3.5 Transport and removal of aerosols

The number of atmospheric aerosols can be reduced by several processes for example coagulation. The most common way that the aerosols can be removed from the atmosphere is by cloud processing and wet deposition by precipitation (see figure 2) *(Chin et al., 2009, Ch. 1).* The efficiency of the particle removal therefore depends on the alignment of aerosols to clouds.

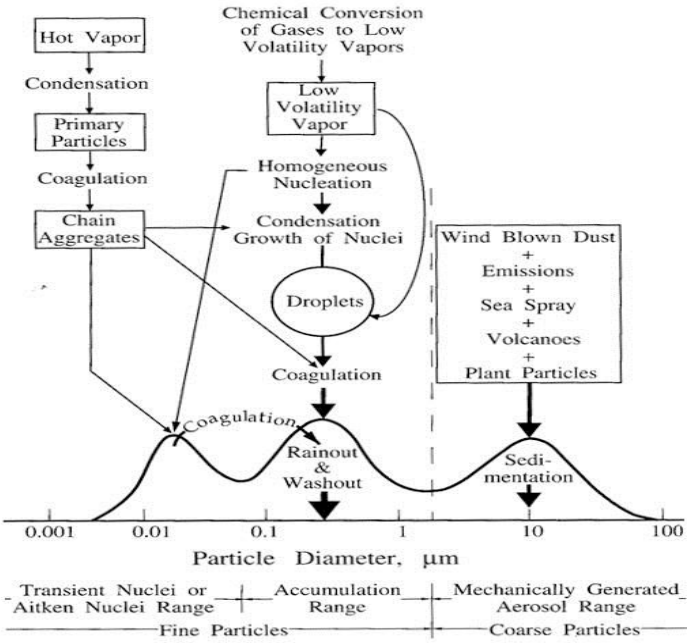


Figure 2. Idealized distribution of particle surface area of an atmospheric aerosol particle modes, formation and removal processes are shown (*Seinfeld et al., 2006, Ch.2)*.

Aerosols can be removed by several other deposition processes for example dry deposition which can be done by gravitational settling that tends to eliminate larger particles (>10,000 nm). Gravitational setting simply means that the particle is falling under gravity; very large particles fall through the troposphere until they strike a surface. Impaction is another form of dry deposition that typically favors particles with a diameter larger than 500 nm.

Coagulation is one way that smaller particles can aggregate with larger ones, resulting in eventual deposition by wet or dry processes. Another way for the aerosols to be removed by rain is through a

wash-out process where the aerosol sticks to an already existing falling cloud droplet and grows larger and gets washed out with the rainfall.

If the aerosols is confide below the clouds that produce rain they can get swept into the rain drop falling down to ground and is deposited with the raindrop. Smaller particles can fall out as raindrops while larger particles could be swept out by the rain. Larger particles have a shorter lifetime and therefore are removed in the matter of hours whereas small particles <20,000 nm could stay in the troposphere for days *(Hinds, 1999, Ch.14).* The removal processes of fine particles (< 1000 nm) are weak, especially particle with a few hundreds nm in diameter, and therefore these particles tend to accumulate. These particles can be removed by rain through coagulation into larger particles *(Hinds, 1999, Ch.14).*

2.4 Particle Size distributions

The size of aerosol particles is an important physical property. The particle size is most often given by one parameter i.e. particle diameter. If the particles are spherically symmetric this is a good approximation to use for calculating different properties of the aerosols. This is often a good approximation for e.g. liquid droplets and some particles formed by condensation and aged particles that have undergone atmospheric processing. Close to a source, the particles are often non-spherical and some corrections are needed to be able to calculate for example the mass size distribution, surface area and other properties. In this thesis all particles are assumed to be spherically symmetric when converting the measured number size distributions to mass or volume.

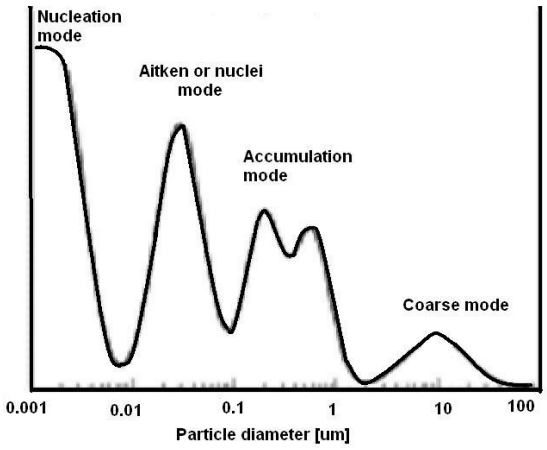


Figure 3. Idealized atmospheric aerosol size distribution. Particles tend to be found in modes that reflect their origin (*McMurry, 2003*).

There can be confusion when discussing particle size and what particles of different size are called; in this thesis the definitions below are used. In general dust and ground material are in micrometer range or larger and fumes and smoke are in the sub micrometer range. The particles that cover a range from 1-30 nm are called ultrafine particles or nucleation mode particles, particles with a diameter < 1000 nm are called fine particles. Very large aerosol particles (large sized particles > 10,000 nm) have a limited stability in the atmosphere *(Hinds, 1999, Ch.4).*

The size of the particles present in aerosols differ of several orders of magnitude from each other. Therefor one normally separates the particle size data into three or four modes (See figure 3), *nucleation* (1–30 nm), *Aitken or nuclei* (30–50 nm), *accumulation* (50-1000 nm) and *coarse* mode

(>1000 nm) *(Andreae et al., 1995).* These modes are often found in literature but can have different size definitions depending on the author.

Fine particles include nucleation, Aitken and accumulation mode particles and can be produced directly from combustion sources. In an urban environment fine particles with a diameter less than 1000 nm tend to be of anthropogenic origin *(Janhäll, 2004, Ch.2*). These fine particles cover one third up to two thirds of the total mass in the atmosphere *(Hinds, 1999, Ch.14).* They have an atmospheric lifetime of days to weeks and can travel distances from hundreds to thousands of kilometers *(Seinfeld et al., 2006, Ch.8).* Most of the particles in an urban atmosphere is in the range less than 100nm and most of the mass is split between the accumulation and coarse mode *(Hinds, 1999, Ch. 14).*

The nucleation mode (1–30 nm) the nucleation mode is not always present because these small particles coagulate rapidly with each other and particles in the accumulation mode. As a consequence these particles grow larger and end up in the accumulation mode and therefore have a very short lifetime as nucleation mode particles. The size distribution of the nucleation mode particles is variable in urban areas and close to a source extremely high concentration of fine particles is found. The concentration decreases rapidly with the distance from the source *(Seinfeld et al., 2006, Ch.8).*

The Aitken mode (30–50 nm) primary consists of particles that are directly emitted to the atmosphere from combustion sources and particles that are formed in the atmosphere by formation processes *(Andreae et al., 2009, Ch.3).* This mode contains a high number of particles. Because of their high number concentration this mode is larger near sources of combustion, for example highways or a as in this study where the traffic is strong.

The accumulation mode (50-1000 nm) consists of fine particles, and includes combustion particles, smog particles and Aitken particles that have coagulated. Because the accumulation mode consists of particles in the visible range (390-750 nm) this mode is the one that is responsible for the reflection of sunlight. This reflection is contributing to climate change.

Accumulation mode particles can remain in the atmosphere for several months and travel long distance because of their relative small size. This is because the particles are large enough to have a low diffusion factor but not large enough to undergo impaction- or sedimentation. Accumulation particles can also have its origin from a condense process of condensation nuclei particles. Since the number concentration of small particles is higher in an urban environment, the accumulation mode tends to be much higher in these areas *(Seinfeld et al., 2006, Ch.8).*

The coarse mode (> 1000 nm) contains the largest particles and tend to be mechanically generated and could for example be sea salt, windblown dust or of anthropogenic origin. Because of their large size coarse mode particles have a shorter lifetime in the atmosphere and tend to settle out or impact on surface in a couple of minutes or days. Despite this, these particles (e.g. mineral dust and sulphate) can travel long distances *(Andreae et al., 2009, Ch.3)* up to tens of kilometers *(Seinfeld et al., 2006, Ch.8).* The most common way to present data from the coarse mode is through volume distributions because they are often few in numbers but have a large mass, while the other smaller particles often are presented by number. In our study coarse mode particles will not be covered because the SMPS instrument does not measure particles larger than 600 nm.

2.4.1 The Lognormal distribution

Atmospheric aerosols particle size can be described by a lognormal distribution function; see equation (1) which is obtained from the normal distribution function by using logarithmic variables. All aerosols do not have the same shape and size and all of them are not spherically symmetric, therefor it is necessary to have a mathematical description in order to describe the size distribution and to be able to analyze the aerosols quantitatively *(Walter, 2005, Ch. 6*). The diameter of aerosols spans over a wide range of magnitude and it is therefore convenient to use a logarithmic scale when calculating size intervals, number distribution, surface area, volume and mass distributions etc.

Equation (1) $$dN = \frac{N}{\sqrt{2\pi}\, d_p \ln \sigma_g} \exp\left[-\frac{(\ln d_p - \ln d_g)^2}{2(\ln \sigma_g)^2}\right] d\ln d_p$$

Where d_p is the particle diameter, d_g = CMD = count number median diameter and the geometric standard deviation σ_g *(Walter, 2005, Ch. 6*). The number distribution function is given by; $N(d_p)=dN/d\log d_p$.

2.5 Meteorology

The meteorology both local and global has a great impact on the outcome of the measurements.

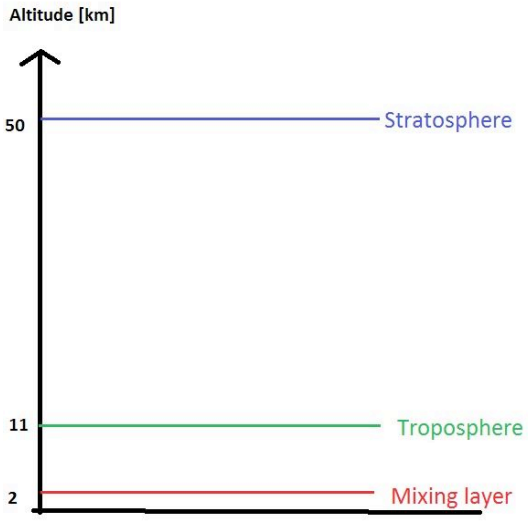
The temperature profile affects the stability of the atmosphere and the vertical air distribution, which has a major impact on the global distribution of aerosols. Solar radiation cause diurnal changes in the stability in the lower atmosphere, caused by radiative cooling during night and heating in daytime. During night in summer time a stable layer (i.e. inversion layer) is established when no clouds aloft and winds are light, as sun rises the radiation increases and heats the ground and air above and the inversion layer is dissolved *(Borgen et al., 2010, Ch. 6).* The inversion layer will be dissolved in a couple of hours and is often dissolved by noon. During the winter the inversion layer can remain all day long when energetic high pressure is present. The inversion layer prevents aerosols from moving vertically and emissions emitted inside the inversion layer will get trapped and high concentrations can occur.

In urban areas the concentration of pollutants from traffic can be much higher in morning rush-hour than in the afternoon, even if the traffic is the same. This is because of the stable inversion layer in the morning and the unstable in the afternoon *(Seinfeld et al., 2006, Ch.16).*

2.5.1 The atmosphere

The vertical distribution of aerosols in the atmosphere (see figure 4) is an important property that determines the lifetime and long-range transport of the aerosol particles. The information of the vertical distribution of aerosols is limited due to technical equipment measurement limitations *(Gong et al., 2009, Ch.4).* However the vertical distribution typically shows an exponential decrease with altitude up to a certain height, depending on climate and emission sources *(Seinfeld et al., 2006, Ch.8*).

During normal circumstances most of the aerosols form a thin haze in the lower atmosphere called the troposphere (<11km) where most of the atmospheric particular mass is confined. Because of seasonal variations aerosols can endure in the atmosphere for much longer in winter than in summer time, which is because of lower humidity during the winter time *(Gong et al., 2009, Ch.4).* In the troposphere the aerosols are washed out of the air by rain typically within a week. About 80 percent of direct emissions are located in the lowest unstable part of the troposphere called the mixing layer or the boundary layer (0-2 km). Therefor the region below 2 km is the most interesting to observe *(Chin et al., 2009, Ch.1).*



Aerosols are also found in the atmosphere just above the troposphere in the region called the stratosphere (11 to 50 km). In this region the aerosols from volcanic eruption can endure for a long time because no cloud formation is present. In the stratosphere the aerosols can remain for a couple of days up to 2 years. Major volcanic eruptions can increase the stratospheric particulate concentration by two orders of magnitude *(Hinds, 1999, Ch. 14).*

Figure 4. Illustration of the earth´s atmosphere.

2.6 Urban traffic

Vehicle exhaust is a major source of particle pollutants that affects the human health in an urban environment. Different types of cars emit different amounts of exhaust with differing composition. Diesel engines exhaust has higher mass of particles than gasoline exhaust. This is mainly due to the combustion process in the engine leading to soot formation, the less clean fuel and that some vehicles still in use have no filtering or catalytic treatment of the emitted gases *(Forsberg et al., 2005).*

The most important factor of exhaust reduction in the atmosphere is the dilution process, i.e. how fast the concentration of the exhausts is reduced in the atmosphere. The reduction rate depends on the volume of the mixing layer; the altitude of the mixing layer can change depending on meteorology. Diesel engines are often used by industries and transportation of different types such as trucks, locomotives etc. *(Lloyd et al., 2001).* In developing countries the exhaust problem is severe as well because old cars from industrialized countries end up here. Old cars often emit larger amounts of exhaust particles than newer cars that have a more advanced technology. Diesel particles are small, they are in the fine particle range i.e. with a diameter of less than 1000 nm (see figure 5), and they often peak in number size distribution between 50-80 nm *(Wehner et al., 2009).* Diesel can give rise not only to primary particles but also a formation of secondary particles.

It is a fact that exposure to diesel exhausts will contribute to a damage of your health; for example this can lead to lung cancer and asthmatic diseases. Diesel particles have an ability to penetrate deep into the human lung and cause great damage. In Denmark the majority of passenger cars have gasoline engines. Gasoline car exhaust peaks in the fine particle range *(Samet, 2005, Ch.22),* and emits far less primary particles than diesel cars. The atmospheric residence time for gasoline is between 12 and 50 days *(Seinfeld et al., 2006, Ch.2)*; this is also valid for diesel particles.

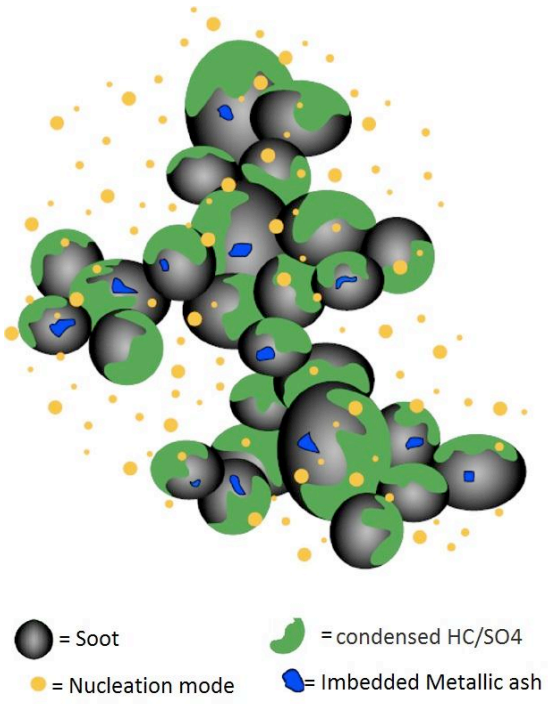


Figure 5. Artist's conception of diesel Particulate Matter (PM)*(Maricq, 2007).*

The major part of the emitted particles from diesel engines is located in the Aitken and the accumulation mode; particles in these modes consist of smaller primary particles that have turned into larger chain like agglomerates.

Close to a roadway high concentration of small fresh combustion particles will be found, resulting in a mass increase in nucleation, Aitken and accumulation mode. Near a source the amount of particles are many and also of small size. Typical, long range transport particles are larger than these original particles near the sources. Part of the particles can also be found in coarse mode due to resuspension of dust particles from the road. The total particle emissions from gasoline cars are up to two orders of magnitude lower than those from diesel cars *(Wehner et al., 2009*).

2.7 Health effects of airborne particles

Air pollutants such as CO, NO_2 and Particulate Matter (PM) (i.e. exhaust from motor vehicles) are believed to all have a large impact on human health. These particles are interesting to study because of their size and chemical composition which also is the main reason why they are hazardous. Particles from vehicle exhaust and other combustion sources contain different organic chemicals that easily can be deposited in the lung. This is also valid for soot particles.

Because these particles are hazardous there are air quality guidelines. In the European region the ambient air quality is regulated by directives from European Parliament and council. There are two directives; one for Particular Matter PM_{10} and one for $PM_{2,5}$(i.e. particle diameter with less than 10 respectively 2.5 µm). The upper assessment threshold is 70 percent of the limit value for PM_{10}; the limit value is 35 µg/m^3 per 24-hours, and shall not be exceeded more than 35 times per year. The lower assessment threshold is 50 percent of the limit value for PM_{10}; the limit value is 25 µg/m^3 per 24-hours, and shall not be exceeded more than 35 times per year. For $PM_{2,5}$ there are no daily threshold values only an annual average (*EUROPEAN PARLIAMENT AND THE COUNCIL, 2008).* The guidelines are set for PM and not for the particle number or surface area, which might be the most important parameter that affects the human health, but this is not yet addressed.

The size of the particles is a direct problem, i.e. the smaller particles the greater damage they will cause. The ultrafine particles can penetrate deep into our lungs, human body and cause great damage. These small particles can cause the following symptoms; problems with respiratory system such as coughing and breathing problems, decreased lung function, asthmatic problems, chronic bronchitis, irregular heartbeats, non-fatal heart attacks and even death for people with already existing heart-/ or lung diseases *(EPA homepage).*

People with already existing problems in their respiratory system, diabetes (like our human exposure group) or heart diseases will be negatively affected when exposed to airborne particles. Today there is not enough evidence of how airborne particles will affect a healthy human, but scientists expect that these small particles will have a long-term effect.

2.7.1 Particle deposition in the human lung

The human body has its own defense system that can handle hazardous particles. Nose hair is the first instance to filter out the largest particles, and then the sticky mucus in the upper respiratory track will capture the smaller particles and handle pollutants in gas form. The cilia in the respiratory tract will move back and forth to transport the pollutants from the throat up to trachea where swallowed. In the alveolar region the main clearing mechanism is the macrophageous, see illustration of deposition in figure 6.

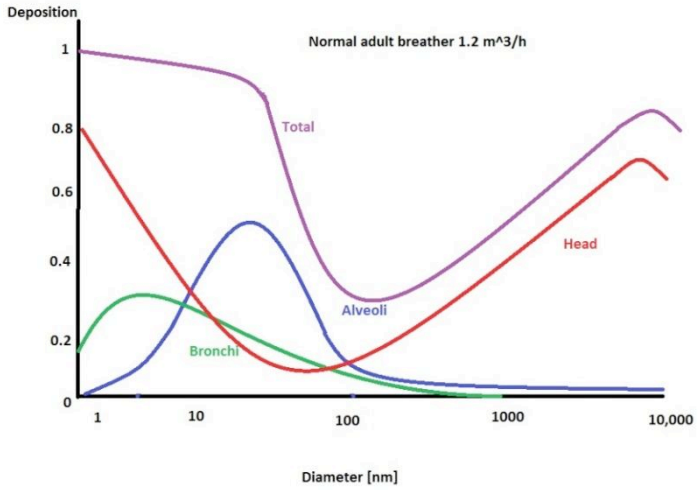


Figure 6. Deposition probability of inhaled particles in the respiratory track according to particle size.

If the human body is exposed to air pollutants in too high concentrations or for longer intervals these natural defense systems will start to break down. This often results in respiratory diseases, asthma, bronchitis etc*. (Miller, 2007, Ch.2)*. There are several reasons why ultrafine particles are very hazardous; some of them are listed here below.

1) Their aerodynamic properties make it possible to enter deeper down in the lung and deposit here. Larger particles (typically > 10,000 nm) will not be able to enter the lung.

2) The reason why these ultrafine particles can enter deeply into the human lung is the fact that these particles are small. In the deep lung where diffusion is an important deposition mechanism they might be deposited. Generally particles < 100 nm have much larger concentrations than particles in coarse mode. This might result in inflammation of the lung.

3) The relative total surface area of ultrafine particles is high compared to coarser particles of the same total mass, and scientists believe that this surface area is connected to free radical activity and oxidative stress (this is believed to cause inflammation in the lung) in the human lung.

4) Airborne particles size are so small, much smaller than the human cellular size, which makes it possible for these particles to enter the human body and even the bloodstream which can cause great danger to the body such as brain damage and heart defects. However, there is still a debate whether translocation of particles are occurring to any large extent.

Another consequence of the ultrafine particles is that diesel particles (mainly soot) are often covered with other toxic substances that are carcinogenic *(WHO, 2006)* and transported down and deposited in the deep lung together with the soot core.

3. Experimental

The particle number size distributions are measured with a Scanning Mobility Particle Sizer (SMPS), with a time resolution of 3 minutes. These measurements span over a size range of particles between 10 nm to about 500 nm.

3.1 Differential Mobility Analyzer (DMA)

The DMA is a device that determines the electrical mobility of particles. In a DMA an electric field is created, the aerosols drift in the DMA according to their electrical mobility. Particle size is then calculated from the mobility distribution. This is based on the physical principle that the ability of a particle to traverse an electric field (electrical mobility) is fundamentally related to particle size; so size calibration is necessary *(Hinds, 1999, Ch.15).* The DMA consists of a cylinder which has a negatively charged rod in the center (see figure 7).

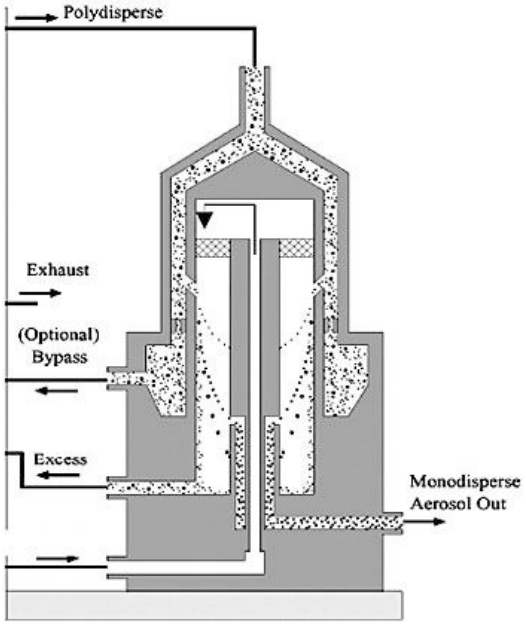


Figure 7. Schematic diagram of a differential mobility analyzer (http://www.azonano.com/article.aspx?ArticleID=1985#_Nano_DMA).

The main part of the DMA consists of particle free air often denoted as “sheath”. Particles are injected into the DMA from the outside of the device edge and then the particles will travel due to an applied electrical field through the sheath, towards the central rod. The velocity of the particles is determined by their electrical mobility. The DMA decides what kind of particles will exit throughout the slit depending on the particles size, charge, flow inside the DMA and the voltage over the central rod *(Flagan, 2005, Ch.18).* In practice it is the electrical field that is varied during the measurements, selecting particles of different electrical mobility in the SMPS (section 3.2).

3.2 Scanning Mobility Particle Sizer (SMPS)

The SMPS instrument is an application of the DMA technique to determine the number size distribution of aerosols, see illustration in figure 8. The SMPS consists of a charger, a DMA and a CPC (denoted UCPC when adjusted to measuring ultrafine particles). In the charger the particles are charged. The DMA is the device that selects the different sizes of particles that we want to measure in the given sample. The CPC is the device that counts the particles in the sample.

The basic principle behind the SMPS is the mechanism of the mobility of charged particles in an electric field. Particles that enter the SMPS system are neutralized i.e. bipolarly charged by a radioactive source. Particles exhibit a Fuchs equilibrium charge distribution which means that all the particles are charged with an equal number of negatively- and positively charged particles. After this the particles enter the DMA where a classification is made according to their electrical mobility; in the DMA one can determine a specific range of mobility of the particles and only these particles will be able to excite throughout the slit of the DMA *(CPS homepage).*

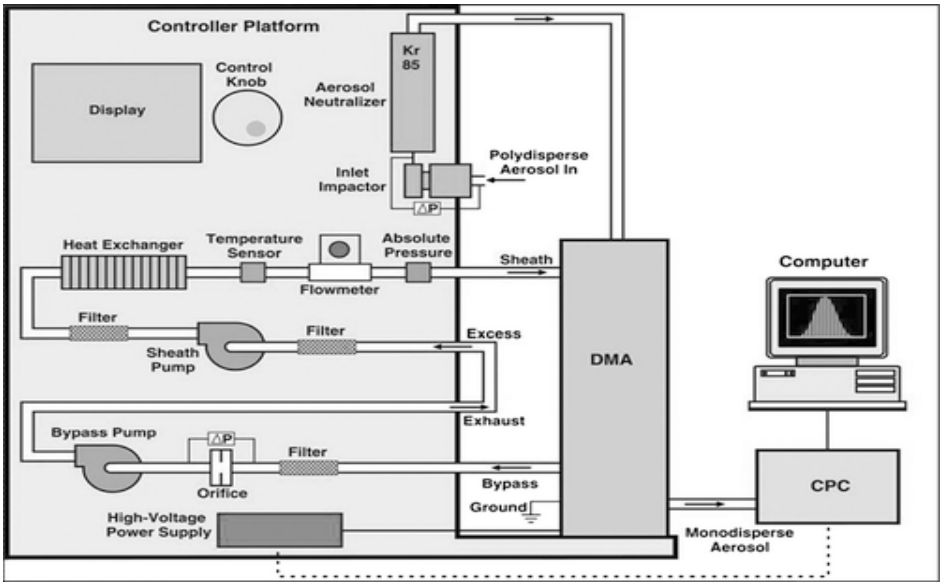


Figure 8. Schematics of a conventional Scanning Mobility Particle Sizer with a long DMA, TSI SMPS Model 3936 (courtesy of TSI, Inc.). The ESC is located on the left from the CPC and a computer *(Hogrefe et al, 2006).*

One important thing about the SMPS is that this device scan the voltage of the rod in the DMA exponentially, resulting in a full size particle distribution typically in the size range on 10-1000 nm. The electric field in the DMA changes continuously therefore this is a fast method to measure and analyze just over one size spectra. The measured particle distributions must be inverted to obtain the actual aerosol size distributions, considering the charge probability, instrumental losses, the DMA transfer function, diffusion broadening and CPC count efficiency etc.

When measuring aerosols in the SMPS system it is important to take the diffusion losses in the sampling line into account. When aerosol particles collide with a surface they adhere due to surface tension, electrostatic forces, and van der Waals forces. The smaller the particles are the more rapidly the diffusion will occur; diffusion is the major transport mechanism for particles smaller than 1000 nm, therefore it is very important to use a diffusion loss algorithm for the ultrafine particles. For

particles larger than 100 nm one can discard this algorithm. If the sampling in the SMPS measurements is made at low flow rates; this will also influence the diffusion losses *(TSI SMPS homepage).*

The charges of the aerosol particles in the SMPS system need a correction formula to calculate the fractions; since there are uncharged, single, and doubly charged particles.

3.3 Ultrafine condensation particle counter (UCPC) and Condensation particle counter (CPC)

The only difference between the UCPC and CPC is their size range; the UCPC can measure smaller particles than the CPC. In the present SMPS system the CPC is used to measure the number concentrations at the selected scanning interval, selected by the DMA. The CPC is used to count the particles in the downstream of the DMA. Particles present in the sample stream serve as condensation nuclei, this condensation is necessary to be able to detect the particles by light scattering since otherwise the particles are too small to scatter light and be detected. Once the condensation begins, particles that are larger than a threshold diameter grow quickly into larger droplets and then pass through an optical detector where they are counted. The UCPC counts the total particle concentration in our set-up *(TSI UCPC homepage).*

3.4 Aerosol Mass Spectrometer (AMS)

The AMS is an instrument that measures different elements. This can be done via two different modes of the device; Particle-Time-of-Flight-mode or Mass spectrum mode. It is most convenient to run these modes one by one with a given time period between them. The Particle Time of Flight mode measures the vacuum aerodynamic diameter of the particles, when they travel from the chopper to the Time-of-Flight- Mass Spectrometer. In the Mass Spectrometer the time of flight of the positive ions are measured; if the ions have different weight they will travel with different velocity, since they will be affected variously by the electrical field due to their mass. Using a calibration software one can determine the specific mass due to their time of flight. This can later on be used to determine what kind of element we are observing *(Nordin et al., 2009).*

3.5 Location of the experimental site

The experimental study was set up close (2 m) to a large road (Øster Søgade) in central Copenhagen near the four lakes, see map in figure 9. Copenhagen is the capital of Denmark with approximately 650,000 inhabitants, located on the east coast (55.6˚N 12.5˚E). Øster Søgade was highly busy by vehicles, during the time of the measurements a redirection of traffic was made due to roadwork; this resulted in higher amount of vehicles passing by the location site. The roadway was during the time estimated to exhibit 50,000 vehicles per every 24 hours *(Copenhagen research description*). Before the roadwork in 2010 the amount of vehicles on Øster Søgade was 17,700 per 24 hours (*København Kommune)*.

Figure 9. Map of Øster Søgade in central Copenhagen (http://nyhederne-dyn.tv2.dk/article.php/id-10009772:dyt-i-trafikken-udl%C3%B8ste-skud.html).

3.5.1 Meteorology

The climate and the meteorology around the location will affect the particles physical and chemical properties. Scandinavia has an Oceanic climate characterized by the moist air from west and the warm Gulf Stream. Copenhagen has a typical maritime climate which means a climate with warm summers and mild winters. Typical wind conditions in Copenhagen are west-/ southwest winds and the average precipitation is minimum during February (35mm) and maximum in August (75mm) *(Borgen et al., 2010, Ch. 6).* During the winter winds from east are more likely, because a thermal high pressure is situated over Siberia.

The temperature profile during the whole period in Copenhagen is shown in figure 10. December until the end of January the temperature is approximately the same. In the end of January the temperature drops below zero.

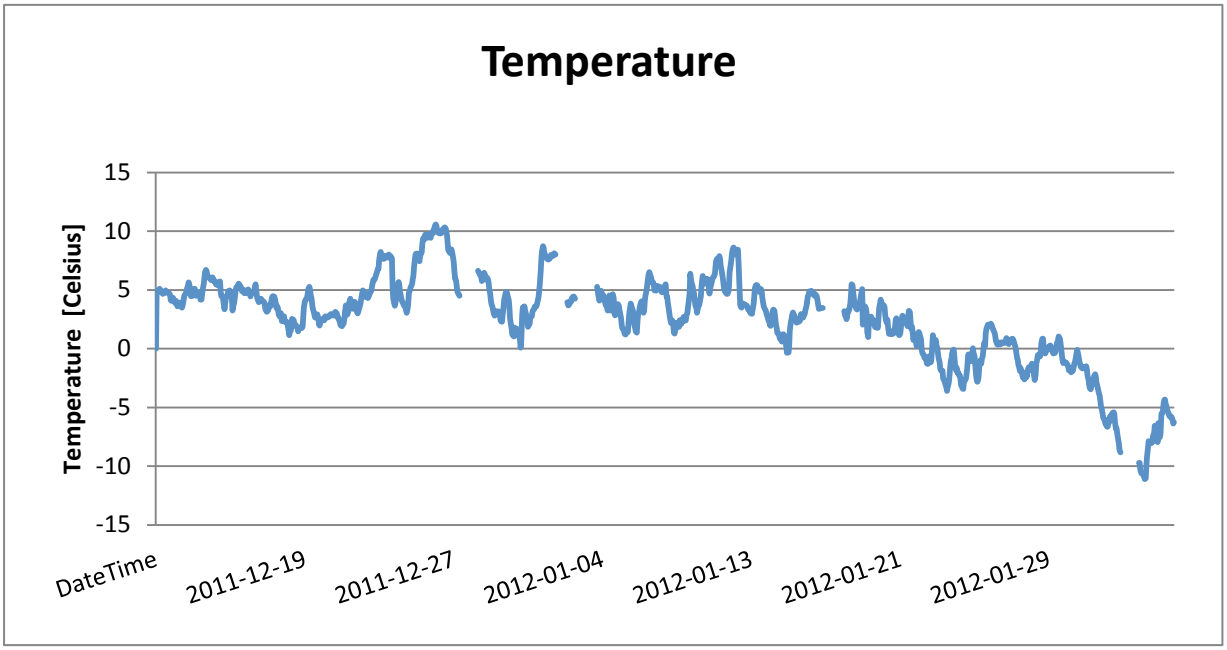
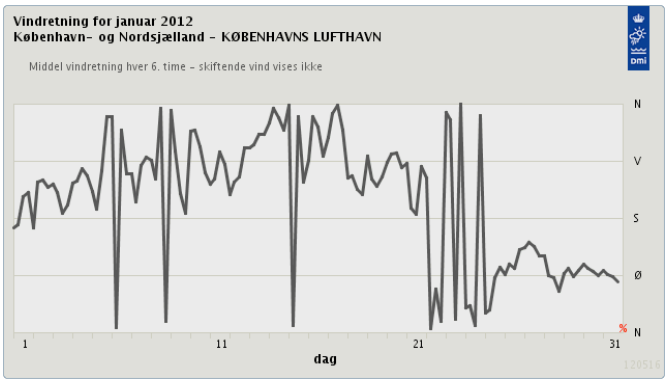


Figure 10. Plot of the temperature profile in Copenhagen December 2011 to February 2012 *(Ketzel, 2012).*



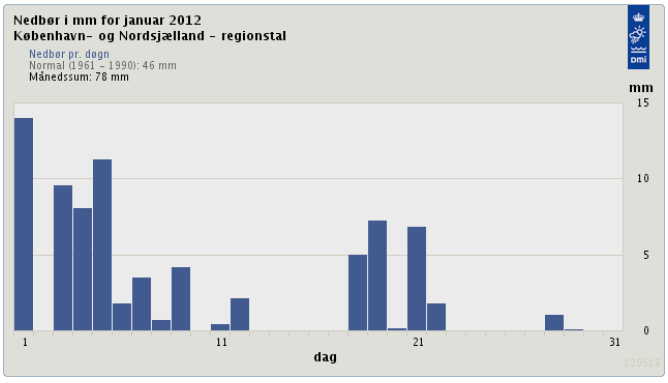


Figure 11. Plots of the wind direction and precipitation in Copenhagen January 2012 (http://www.dmi.dk/dmi/vejrarkiv?region=7&year=2011&month=12).

The local wind direction and precipitation in January in Copenhagen is presented in figure 11. When we have western/northern wind the wind blows perpendicular to the building were the measuring equipment is situated.

In the beginning of January the amount of precipitation was larger than in the end. If the aerosols confide below the clouds that produce rain they can get swept into the rain drop falling down to ground and deposited with the raindrop. Smaller particles can fall out as raindrops while larger particles could be swept out by the rain

4. Experimental Measurement Analysis

4.1 Trajectories

Trajectories are a determination process; the trajectories are an estimation of where a certain air parcel comes from and how it has traveled before reaching an experimental site see figure 12. It is possible to determine the parcels origin; i.e. to track this parcel back in time, it is also possible to try to predict how this specific parcel will move and where it will be located in the future. The trajectories are an outcome from meteorological data from all over the globe. At Air Resource Laboratory (ARL) all the meteorological data needed to produce trajectories can be downloaded.

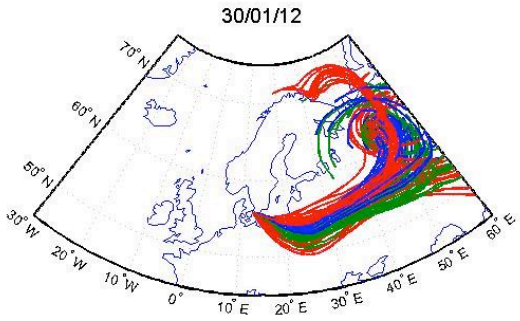


Figure 12. Trajectories of three different heights (100 m=blue, 500 m=green, 3000 m=red) from Copenhagen 5 days back in time.

4.2 Data analysis

This thesis focuses on the SMPS which is one of many instruments in the human exposure study made in Copenhagen. Two different experimental setups were used; one were the SMPS was in front of the other instruments (set up 1) and one were the SMPS was after the other instruments (set up 2). When the SMPS were in the first experimental setup all the data could be used but in the second setup there were difficulties in the determination of what data that could be used in our analysis.

The health study was made on both indoor and outdoor air. When human test group was exposed to indoor air in the exposure room, the data had to be neglected; this because our analysis only covers outdoor air. Most of the indoor exposures were made on weekdays in January which means that much data in January had to be neglected.

There was an offset in the SMPS instrument between 20 /12/2011 and 4/1/2012, therefore a new inversion (made in LABWIEV, a programing language) of the data had to be done. The inversion was necessary because all the particle diameters were wrong; the offset caused greater error on smaller particles. A logbook over all the measurements was recorded continuously, and from this we could determine what data that could be used for our analysis.

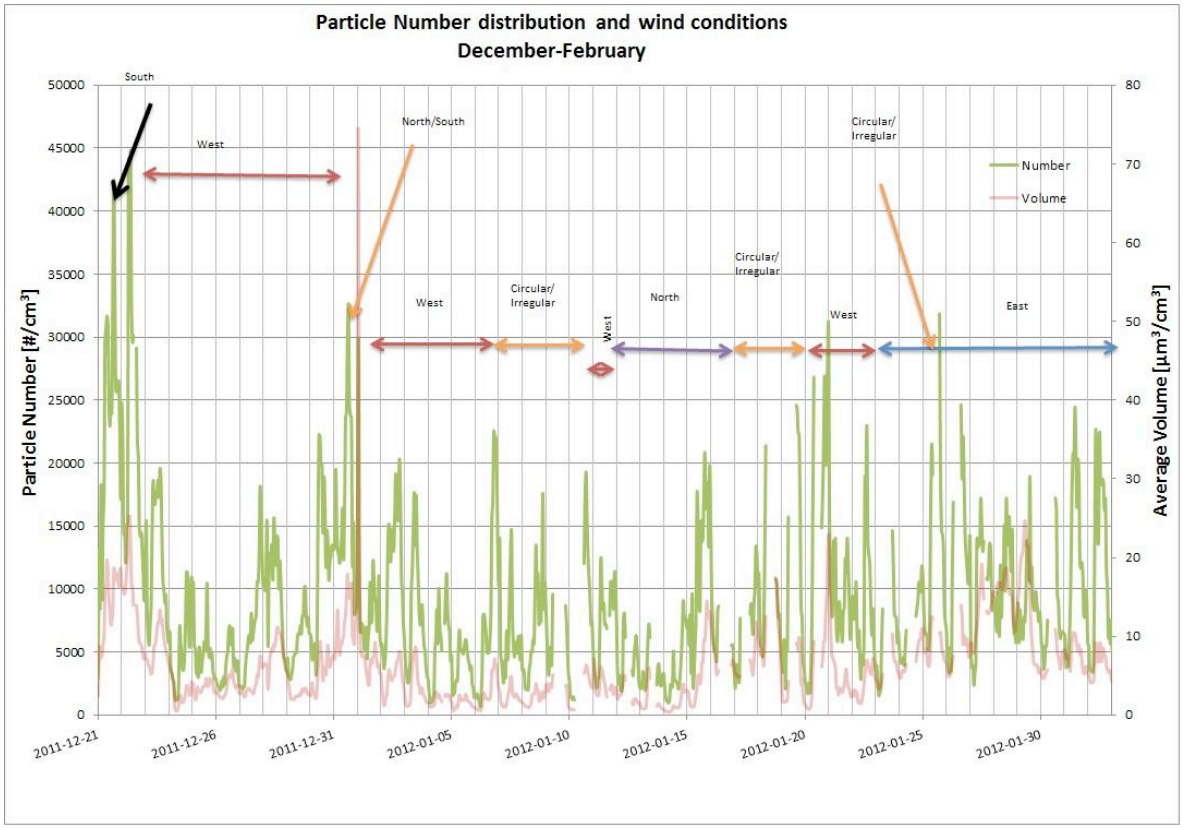
4.3 Data interpretation

In this thesis two computer programs where used; MATLAB and HYSPLIT. Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) Model is a program that handles the meteorological data downloaded on (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1); the HYSPLIT model is a complete system for computing simple air parcel trajectories to complex dispersion and deposition simulations. HYSPLIT gives us trajectories that can be used later on in the determination process in where the air parcel comes from. The air in the parcel will absorb particles along the path.

To determine and plot the trajectories a program written in Matrix Laboratory (MATLAB) was used. In this program wanted coordinates could be chosen (in this thesis Copenhagen), and the trajectories could be chosen at three different heights (100, 500, 3000 meters). The two most important heights of trajectories when doing aerosol analysis are the two lowest heights, that is 100, 500 meters, this because the 3000 meter height is located above the mixing layer. Since 80 percent of direct emission is located below the mixing layer the 3000 m height is not so interesting to study. The program plots an image over the trajectories of the three different heights on a map; where coordinates on the map can be chosen so that width and heights on the map can be determined. Another MATLAB program was used to plot the SMPS data in three dimensions; where the lognormal distribution was used ($dN/dlogd_p$). The 3D-plots were made because it is easier to study particle concentrations in a logarithmic scale.

5. Result and discussion

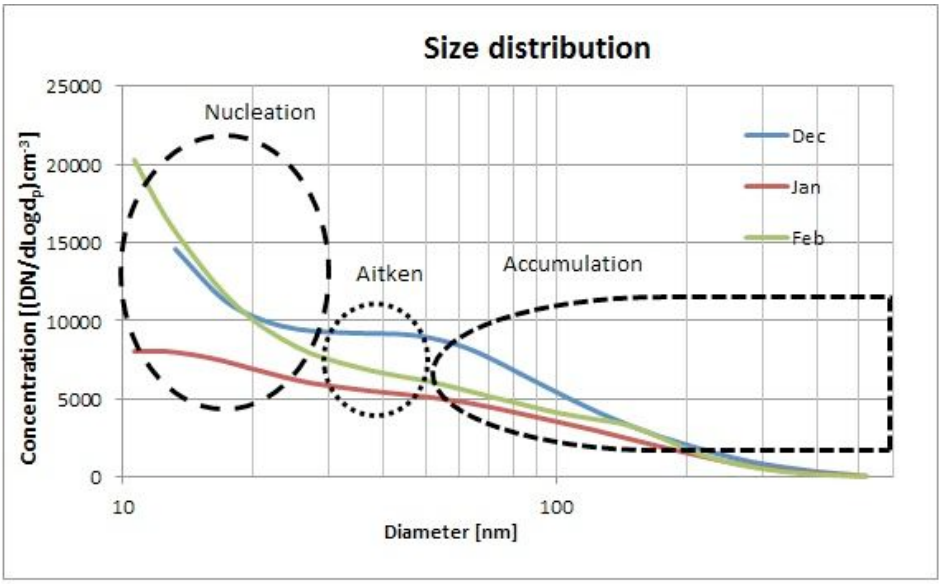
5.1 Overview of the measurement period December 2011-February 2012



This is an overview of volume, particle number and wind conditions during the measuring period 21/12/2011 – 2/2/2012 in Copenhagen. An increase in particle number is seen when wind conditions changes. A correlation exists between an increased volume, wind change and decreasing temperature in the end of January is observed, see figure 10 for temperature. During the whole measuring period the average particle number is 9328 /cm^3, for averages over respectively month see table 2 below. The average particle numbers in each month do not differ so much from each other, even though we have two large but short peaks in December with over 40,000 particles per cubic centimeter. The average in December is between 21/12/2011-31/12/2011, the average in January is over the whole month, in February the average is only for two days.

Month	Number (/cm^3)
December	11299
January	8361
February	11425

Table 2. Average particle number per cubic centimeter.



This plot shows an average size distribution of the particles every month. Again, keep in mind that February is only averaged over two days. All three months most of the particles are in the range between 10-100 nm. This region is interesting to study because it is the most hazardous to human health, this because many particles will deposit in the alveolar part of the lung (see figure 6). In December the Aitken mode is more distinct than in January and February. We can see the nucleation and accumulation mode in all three months. In December the accumulation mode, expected to consist of soot from diesel exhaust (50-80 nm) peaks at 60 nm and is more distinct. The coarse mode cannot be seen because the SMPS measures particles only in range 10-500 nm.

5.2 Particle traffic pattern

Our measurements show an increase in concentration during daytime from about 5 am to 8 pm. We cannot see a typical rush hour traffic pattern in concentration during weekdays. From reading other studies, we expected a diurnal pattern in concentration, increasing during rush hours in both morning and afternoon than observed. Instead the concentrations during day time are smeared out. This could be a result of constant traffic during the day; another possible explanation could be that Øster Søgade has an open street space with the four lakes nearby see the map in figure 9. The open space contributes to an increase in mixing of the aerosols. Because of the data lost in January due to the human exposure study (instruments sampling from exposure room instead of from the street) many weekdays are difficult to analyze and therefor a typical traffic pattern is hard to determine. Since Øster Søgade is situated in the central Copenhagen truck traffic is not so heavy, and therefor the diesel exhaust is less than near highways.

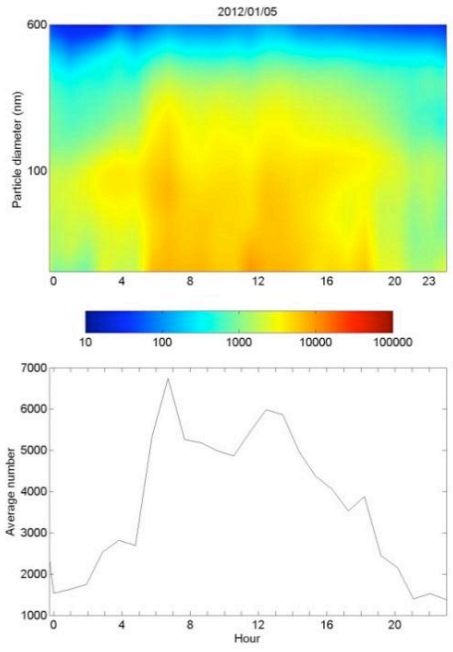
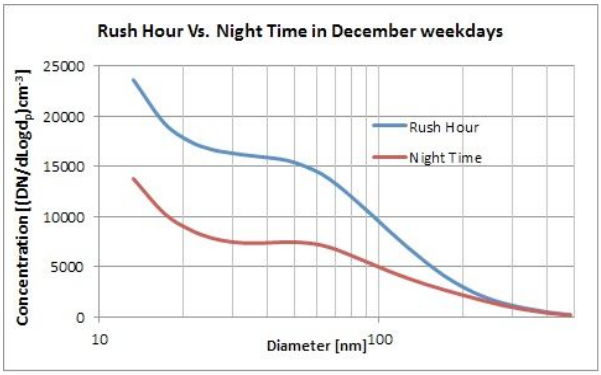
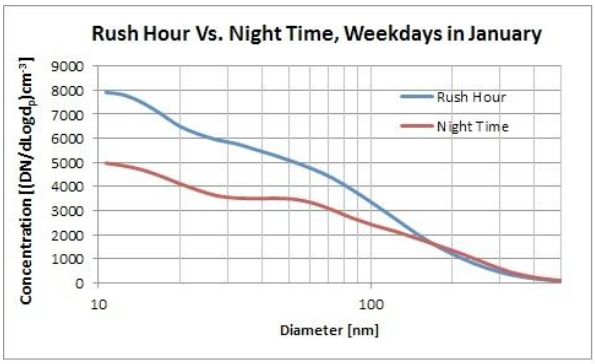


Illustration of the particle concentration and average number during a weekday in January.

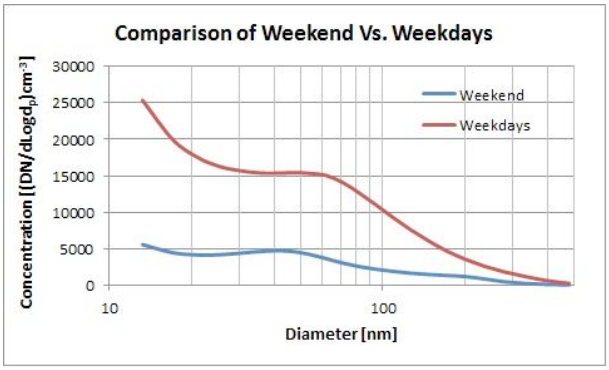
5.2.1 Weekly variations

In the two plots rush hours include hours between 06.00 and 10.00 and night time is between 23.00-05.00. In the December plot weekdays include 21/12-23/12, 27/12-30/12, in January weekdays include 4/1 – 31/1 2012. The two curvatures in each plot have approximately the same shape with the only difference that the concentration is almost twice as large during rush hour. The reason why to compare night time and rush hour is because we want to see if we could determine the background aerosols at the location site. We can see a small increase in particle size during night time. We expected that the aerosol found during night time would be relatively more influenced by long range transport particles than during day time, as well as slightly more aged particles, and that this would result in larger particles in the night time curvature. It is a similar curvature during night time and rush hours, but we can see an increase of particles larger than 150 nm during night time in January. This could be a result of similar emission sources day- and night time, but of lower intensity, i.e. the traffic is still present but not as strong during night. It could also be the fact that the aging process of the fresh combustion emission particles goes slowly, and that the particles produced during daytime are trapped in the inversion layer during the night. The concentration in December is almost three times larger than in January, it is hard to say why this is so, but the traffic pattern should be the same during these two months and the temperature is approximately the same. The winds in December are south and west whereas they in January are west, east, north and circular. From this we cannot draw any specific conclusions. Both the local and global winds follow each, so this should not be a result from differing wind conditions.

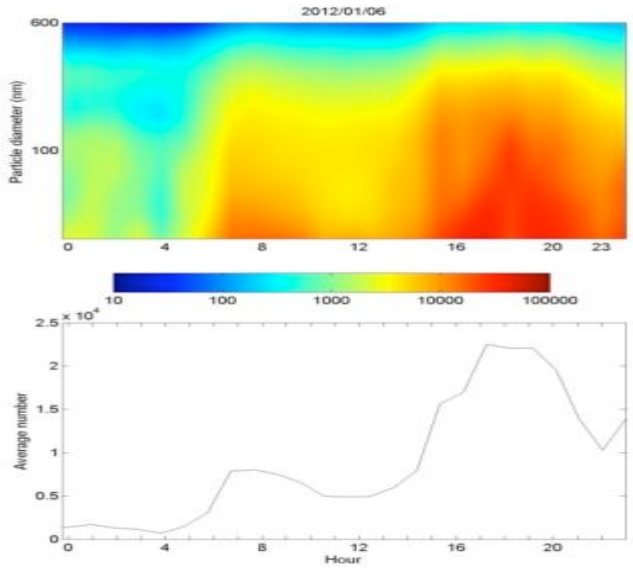




Plot over the weekdays 21-23 December and the weekend 24-25 in December 2011. During weekdays the concentration of particles are higher and a more distinct nucleation and Aitken mode can be seen. We can see a peak in the Aitken mode at somewhat lower size (30-50 nm) in weekends. Decreasing concentration during the weekend can be a result from less traffic and industry emissions. Since the traffic is less during weekends the diesel exhaust is decreased and therefore the Aitken mode (soot from diesel exhaust) is not as distinct. The plot shows the week of Christmas and maybe the traffic pattern is not representative to normal winter days, but since we have much loss in data in January this was the best week to compare weekdays versus weekend.

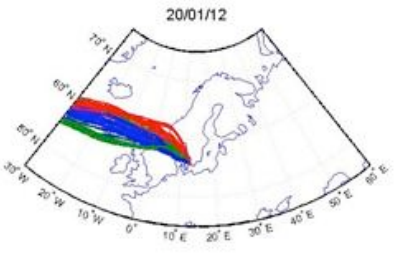


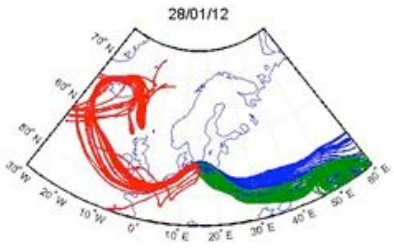
5.3 Wind conditions

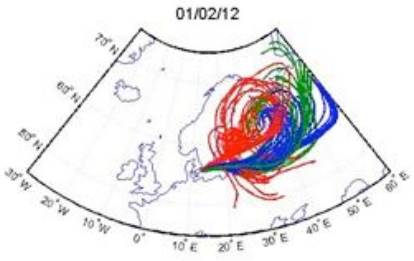


In Copenhagen the typical wind condition is western winds. During our measuring period the wind changes direction many times which is shown in section 5.1 with some typical periods. We can see a correlation between wind change and an increase in particle concentration, illustrated in the 3D-plot on the left hand side.

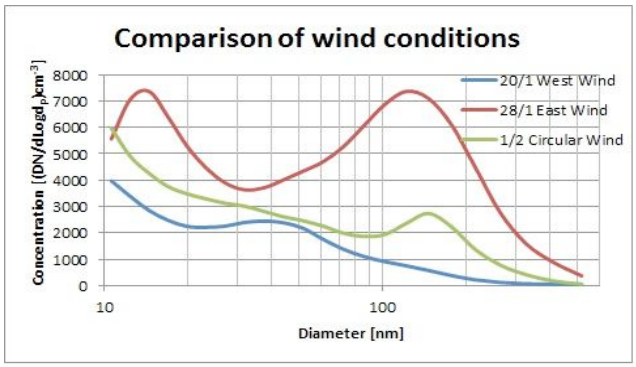
From our trajectories we choose three different days with distinct wind types; western, eastern and circular winds. The chosen days were 20/1, 28/1 and 2/2 2012, see the specific trajectories below. The blue lines represent an altitude of 100 meters, green lines 500 meters and the red lines 3000 meters. We can see that the green and blue lines follow each other but the red lines do not, the red lines are located over the mixing layer and are not so interesting for our study since 80 percent of direct emission is located below the mixing layer.



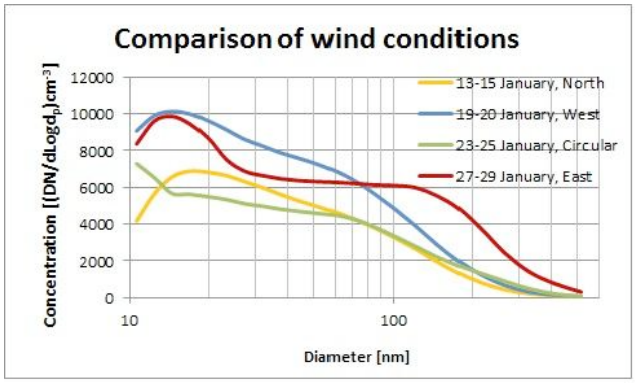




A correlation between wind types and size distribution can be observed via this plot and the trajectories. The eastern wind shows a significant nucleation and accumulation mode, which means many small fresh combustion particles, and larger long range particles. In the western wind curvature we cannot see any distinct contribution from long range transport, but the nucleation and Aitken mode is present. During the occasion with circular wind a nucleation mode and a distinct accumulation mode is observed.



The eastern wind (28/1) is a Saturday which means less traffic. Still we can see a distinct nucleation mode; since we are measuring in an urban environment located 2 meters from the emission source i.e. the road. The local winds are also eastern, which means that the winds have travelled over Copenhagen city and might have brought some fresh nucleation particles along. It could also be that gaseous species from east condense over Copenhagen and small particles fall out. We expected that northern or western wind types would be the most favorable because they blow perpendicular to our location site, bringing fresh particles from the roadside, since these winds would then blow perpendicular to the building. However, we can see that every time we have eastern local winds the nucleation mode is more distinct and increased. When the wind blows from the east we can also see an increase in larger particles which is correlated to long range transport. From the illustration in figure 1 we can see a lot of industries with strong emissions located mostly in Poland and Germany. Since the wind was blowing from the east this could be related.



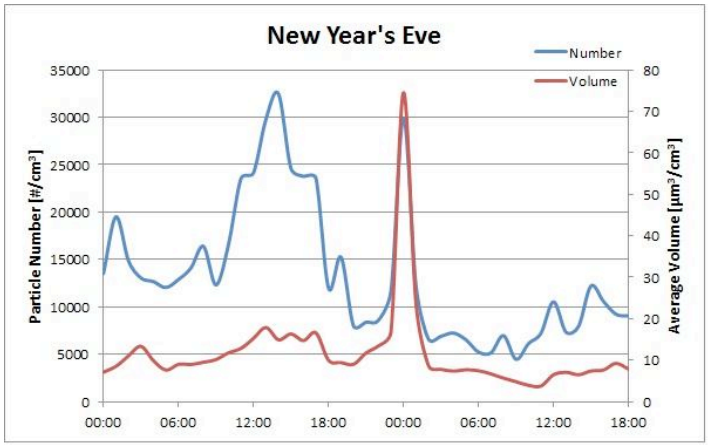
The circular wind shows many small particles in the nucleation mode, but also shows an accumulation mode. Here the local winds are easterly which can result in an increased nucleation mode because it then blows perpendicular to our experimental site.

The western wind shows a clear nucleation mode and Aitken mode, which is related to the traffic exhaust and particles that are formed in the atmosphere by formation processes. What we do not see here is the accumulation mode. Since the illustration in figure 1 shows less emission sources in the west, the long range transport is expected to be lower than from example the east. To see if the correlation between winds and size distribution is valid over longer time, we made another comparison, shown in the illustration “Comparison of wind conditions”.

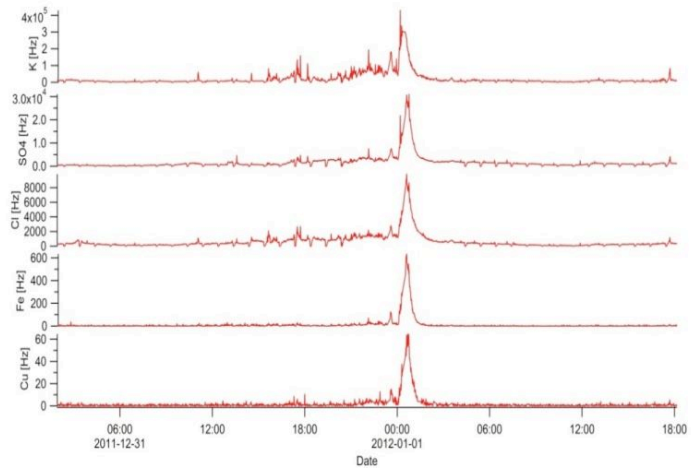
If we had the time we could have done a chemical analysis with the AMS of the particles that we measured with the SMPS, and through that determine if the particles are long range or not.

5.4 New Year's Eve

During New Year's Eve we can see a significant increase of particles; this increase is correlated to the fireworks projected during midnight, shown in the number size distribution plot below. We can see an increase in both particle number and volume during midnight since the particles are large the volume increases. Because this increase of particles is in the accumulation mode, the first assumption would be that this comes from long range transport, but since we know that the four lakes is a place where people project a lot of fireworks during midnight, we can assume that this is not long range particles. In the particle number and volume plot we see the distribution 24 hours before midnight and 18 hours after. The peak in number concentration around noon is larger than midnight. This could also be result from heavy traffic. Another explanation would be some early projected fireworks near the four lakes, but this is likely not the case since the pattern of chemical composition at New Year found with the AMS, and the different size distribution from the fireworks (from SMPS at 23-00) is different.



The AMS does not measure particle sizes under 70 nm very efficiently, but this is not a problem during New Years Eve since we have large sized particles. The plot below shows a selection of substances typically found in fireworks; there are many other substances from fireworks measured by the AMS. The *y*-axis of the plots of the substances is individual and therefore we cannot compare them to each other. We can still see that during midnight we have a clear increase of heavy metals, and that the chemical fingerprint is very specific during this hour.



6. Conclusions

In the overview section 5.1 we can see that the average particle number distribution does not differ so much between the three months, even though we have two peaks over 40,000 particles/cm^3 in December. In the end of January the total particle volume increases and we can see a correlation with decreasing temperature and winds from East. We can observe larger amount of particles every time the wind changes direction.

During the measurement period we have the highest concentration of particles in the range between 10-100 nm, which corresponds well with theory. The particles in this region are the most hazardous to human health, since they are small and deposit in the lung. This is favorable when doing human exposure studies.

We can see the nucleation and accumulation mode in all three months. In December the Aitken mode is more distinct than in January and February. In December the soot/diesel (50-80 nm) peaks at 60 nm and is more distinct. The coarse mode cannot be seen because the SMPS measures particles in range 10-500 nm only.

The diurnal pattern in concentration due to traffic is not as distinct as we thought during rush hours; instead we have an increase in concentration during daytime. This can be due to road works and the local topography with the open street space and the four lakes, which contribute to a large mixing volume.

In December and January the particle traffic pattern has similar curvature, but December has three times larger concentration of particles. We can see a small increase in particle size during night time. Overall it is the same curvature during night time and rush hours, which probably is a result of same emission sources day- and night time, i.e. the traffic is still present but not as strong during night. It could also be the fact that the aging process of the fresh combustion emission particles goes slowly.

In the comparison of weekdays versus weekend we can see that the concentration during weekdays is higher and has a more distinct nucleation and soot/diesel mode. During the weekend the concentration decreases and a typical peak in Aitken mode is observed.

When we see an increase of particles during night time it can be correlated to change in wind directions. A correlation between local and global eastern wind conditions and long range transport; during these conditions a distinct nucleation mode is present. Local eastern winds mean that the winds have travelled over Copenhagen city and might have brought some fresh nucleation particles along. Furthermore, during the occasion with eastern winds we can see an increase in larger particles, which is correlated to long range transport. We can see a lot of industries with strong emissions located in Poland, Germany where the wind has travelled over.

At New Year's Eve an increase in particle number, size and volume is observed during midnight. The particles are typically larger than a normal day. Since the particles are of large size the deposited fraction in the lungs is lower than for traffic.

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