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# The Metal Content in Airborne Particles in Landskrona

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### Abstract

Landskrona has for a long time had problems with high concentrations of Pb and other heavy metals in the air. Measurements investigating the content of airborne particles have been performed in 1977, 1988, 2003 and 2008 and show an influence from heavy industries, traffic in town and transboundary pollution. To investigate the air quality in terms of heavy metals in Landskrona today, the metal concentrations in atmospheric aerosol particles (airborne particles) will be determined and the sources contributing to them will be assessed.

Aerosol sampling was performed with stacked filter units at two locations, a small harbour (Lundåkrahamnen) and at the town hall (Stadshuset) in Landskrona between 15 February and 23 March, 2017. Aerosol sampling occurred daily on filters and the elemental concentrations were determined by particle induced X-ray emission (PIXE). The elemental analysis works for inorganic elements from Sodium to Uranium, for which some can be detected in concentrations around  $1 \text{ ng}/\text{m}^3$  and others significantly lower. The elemental concentrations were modelled with Positive Matrix Factorization (PMF), a source-receptor model, from which the results provide and describe source profiles and source contributions in absolute levels ( $\text{ng}/\text{m}^3$ ). The sources were identified by analysing characteristic elements and meteorology data.

Compared to the measurements in 2008, the average concentrations for most elements are higher. In the previous campaigns, basically all elements decreased gradually from 1977 to 2008 but in 2017 the downward trend was not observed. Increased average concentrations for both lead (Pb) and zinc (Zn) were observed at Lundåkrahamnen however not at Stadshuset. The average concentration of chlorine (Cl) showed a great increase at both Lundåkrahamnen and Stadshuset compared to previous years and is explained by more winds from the sea and higher wind speeds.

The results from the PMF shows that there are 6 sources contributing to the elemental concentrations in the atmospheric aerosols in Landskrona. The sources are: a soil dust source, a sea spray source, a combustion source, a lead source, a copper and zinc source and a long-range transport source. The long-range transport source was only of importance for the elemental concentrations in fine particles and contribute with a part of the lead concentration. The lead source and copper/zinc source originate both from the local industrial area, where the lead source is likely to be linked to Boliden Bergsöe AB and the copper/zinc source to the shipyard. The combustion source is highly affected by emissions from marine traffic in Öresund.

The aerosol sampling in Landskrona shows elevated concentrations of lead, zinc and copper in the air. Local sources located in the industrial area are the main contributors to the heavy metal concentrations. Although the average concentrations are not alarming and well below the Environmental quality standards (EQS), the risks for chronic lead exposures should be minimised by identifying important processes giving rise to particulate emissions or by applying stricter EQS.

## Acknowledgements

I would like to thank my thesis supervisor Adam Kristensson for all the help and support he offered me during this project. Whenever I encountered a problem he was there with his patience and knowledge to help me find a solution. Adam helped me with insightful comments and steered me in the right direction completing this project. I am beyond grateful for the help he offered when installing and executing the measurements in Landskrona.

I also would like to thank my assistant supervisors. Patrik Nilsson was always there to help me with useful tips and the practical experience when building the experimental setup. Jan Pallon passionately contributed with aid and guidance and provided the PIXE results. Pontus Roldin offered advice and arguments about the HYSPLIT model.

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# 1 Introduction

Airborne particles, also called aerosols, have a great impact on both climate and health. In 2012 it was estimated that outdoor aerosols cause 3 million premature deaths worldwide per year.[1] Exposure to aerosols, especially fine particles, at high concentrations has showed to increase outbreaks of cancer.[2] Studies show that particle characteristics such as shape, chemical composition and size are the defining factors in how human health is affected. The toxicity of aerosols is highly defined by the metals they contain, where some cause inflammatory effects on the lungs.[3] Aerosol pollution becomes a more serious issue when heavy metals come in contact with respirable particles and increase lung injuries. Natural heavy metal emissions, from for example forest fires or minerals in the Earth's crust, will form and stick to coarse particles while human-made heavy metals distribute among the fine particles.[4]

Besides from having an effect on health, anthropogenic aerosols with different compositions cause environmental effects. A direct effect caused by aerosols is the absorption or reflection of sunlight, which warms or cools planet Earth. Indirect effects occur when aerosol particles serve as condensation nuclei for cloud formation which typically cause cooling. The environmental effects occur when anthropogenic aerosols take part in the direct and indirect effects of natural aerosols.[5]

To address the problems caused by aerosols, an environmental quality standard for outdoor air has been set with defined limit values for different aerosol compositions, which includes certain heavy metals, and particulate matter with particle diameters up to 10  $\mu\text{m}$  for PM10 and up to 2.5  $\mu\text{m}$  for PM2.5.[3] The municipalities are responsible for monitoring the compliance of the environmental quality standards and act with proper measures when needed.

## 1.1 Research objective

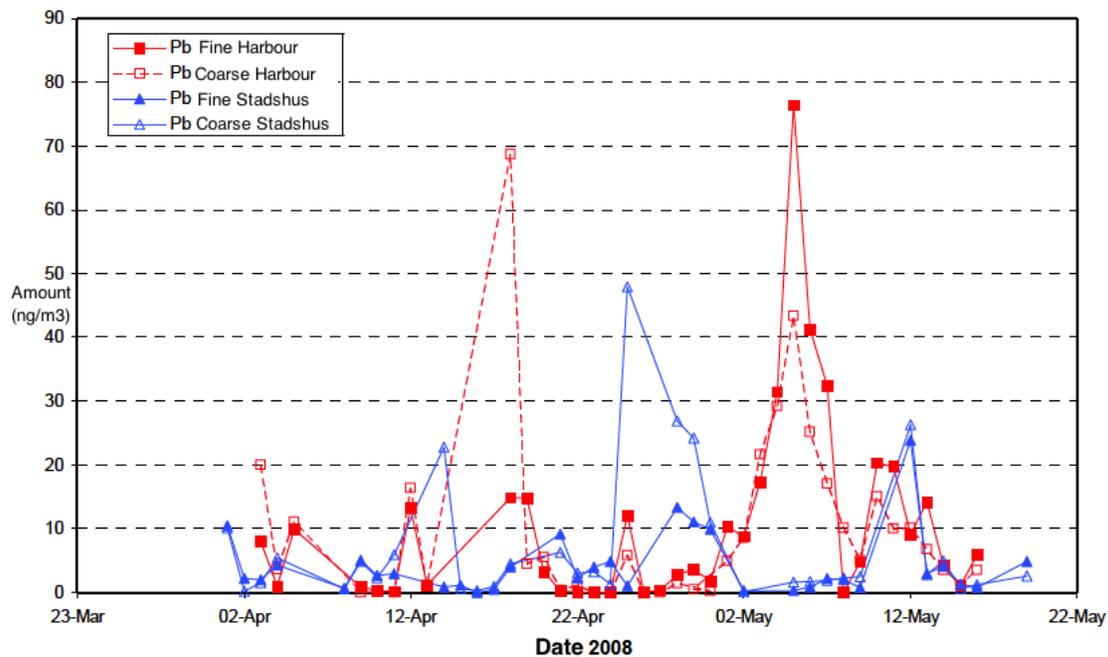
The main research objective is to study and determine the concentration of heavy metals in airborne particles in Landskrona. A specific objective is to asses the sources contributing to the heavy metal concentrations in airborne particles in Landskrona. Similar aerosol measurement campaigns with the same goals have been executed around every ten years with a start in 1977 and continued in 1988, 2003 and 2008. The current measurement campaign will serve as a continuation on the previous aerosol measurements. The Environmental Health Department in Landskrona ordered new measurements in a collaboration with the Aerosol group of Nuclear Physics at the Faculty of Engineering at Lunds University. The measurements should work as a control of imposed improvements, investigating the environmental condition of the air, and if necessary support changes in emission limits. A coordinated recipient monitoring programme including i.a. Boliden Bergsöe AB and Befesa Scandust AB, was created to secure the execution of necessary measurements and to reduce the cost for the involved parties by sharing the overall cost for a measurement. In the programme it was determined to from now on execute measurements on airborne particles every fifth year.

## 2 Background

### 2.1 Air quality in Landskrona

In the city of Landskrona, measurements on the content of airborne particles has been performed during the years 1977, 1988, 2003, and 2008, showing a significant influence from the heavy industries, traffic in the town but also from transboundary pollutions. Particles containing high amounts of heavy metals have been a reoccurring problem in Landskrona and could cause both health and climate effects to the population.[6]

From previous measurements, especially the most recent in 2008, it can be concluded that the concentration of heavy metals, such as lead and zinc, still show elevated values compared to the measured levels in other cities. In figure 1 both the concentration of lead in coarse (PM10) and fine (PM2.5) particles has been presented for the measurement station at the harbour, which lies close to the industries, and the station called Stadshus which lies downtown. The same measurement stations will also be used in this project. The figure shows peaks in the concentration of lead at different times for the different locations which could be due to varying wind directions where a certain wind direction corresponds to a possible lead source. The daily values of these peaks are below allowed environmental quality standards but are as mentioned elevated.[6]



**Figure 1:** The lead concentrations for the coarse and fine fraction measured at Lundåkrahamnen and Stadshuset in Landskrona in 2008.[6]

A more recent study from 2015 shows high concentrations of metal particles in falling dust, particles that sediment on the ground, in Landskrona. The falling dust contains especially high concentrations of lead and zinc. Hence it is likely that high concentrations of these metals will be measured in the current 2017 campaign on airborne particles in Landskrona.[7]

## 2.2 Aerosol sources in Landskrona

The aerosol sources in Landskrona can be divided in three main categories, local industries, urban and transboundary sources. Landskrona is a harbour city located in the south of Sweden with important heavy industries that emit relatively high concentrations of aerosol particles. Boliden Bergsöe AB is an industry that recycles metals from approximately four million recycled lead batteries per year. This produces approximately 45 000 tonnes of lead alloys per year from which 60% is reused in battery production and the remainder in other applications.[8] Recent measurements from 2015 in the vicinity of Boliden Bergsöe AB show high amounts of lead, zinc and cadmium in falling dust.[7]

Another heavy metal recycling industry in Landskrona is BEFESA Scandust AB where valuable metals such as chromium, nickel, molybdenum, manganese and iron are recovered from waste dust which are filtered out during the production of stainless steel. By using a plasma technology, the waste dust can be recycled and reused in the production process of stainless steel. BEFESA Scandust AB has a yearly capacity of receiving 65 000 tonnes of dust. The amount of heavy metals in the dust has been lowered over the years from 16% to 11%.[9] In the vicinity of Scandust, high amounts of lead, chromium, nickel and zinc have been measured during 2015 in falling dust.[7]

Urban sources in Landskrona are traffic emissions in the form of exhaust particles and tire and road wear particles. Traffic fumes contribute with ultrafine particles. The tire and road wear particles produced by local traffic are resuspended and constitute to a large portion of the PM10 amount. Combustion processes for house heating contribute to the emission of ultrafine particles in cities. As Landskrona is a harbour city ship exhaust is expected to contribute to the Landskrona particles.

The last category is transboundary sources, which includes long range transported air pollution from sources in continental Europe. A variety of long-range transported particles can contribute to high particle concentrations, for example sea spray particles, dust particles, fossil fuel combustion, biorganic particles and other sources.

## 2.3 Earlier aerosol measurements in Landskrona

One of the first measurements executed in Landskrona on the concentration of metals in airborne particles showed similar amounts as measured in Malmö except for the amount of lead which was twice as high. The sources responsible for the high amounts of lead in the aerosols were found to be Boliden Bergsöe AB and the traffic in town.[10] When at a later stage, wind directions were accounted for, it was proven that the air quality in downtown Landskrona was highly affected by the industrial area located to the south of Landskrona. [11] Measurements in 2008 showed a descending trend for the concentrations of heavy metals in aerosols and on average the amounts corresponds to the amounts measured in background air in south Sweden. However occasionally the concentration of lead, zinc, chromium and nickel are still showing high concentrations and are traced to local sources such as BEFESA Scandust AB and Boliden Bergsöe AB. [6] The results from recent measurements performed in Landskrona do not show any clear long-term trends on metal concentrations except for the concentrations of lead which have decreased in 2014 and 2015 compared to previous years.[7]

## 3 Theory

### 3.1 Atmospheric aerosols

#### 3.1.1 Definitions

Aerosols are colloidal suspensions of solid and liquid particles dispersed in air. In other words, the air that we breath is an aerosol. The smallest suspended particles are in the range of a small cluster made up from several molecules (1 nm). The largest suspended particles classified as aerosols have a size of 100  $\mu\text{m}$  and are limited by sedimentation speeds where larger sizes makes the particles disappear from the suspension in a rapid rate . Aerosols produced in a laboratory can have uniform sizes and are classified as monodisperse aerosols while most outdoor aerosols are polydisperse and contain a range of different particle sizes.[12] There are several ways to measure particle concentration but the most common ways are measuring the mass per unit volume or the number of particles per unit volume.

Aerosols particles, especially the solid ones have very often irregular shapes that require the use of an equivalent diameter for comparison with spherical particles with similar physical properties. For example one can use the aerodynamic diameter of an irregular shaped particle corresponds to the size of a spherical particle with the same sedimentation speed and a density of  $1\text{g}/\text{cm}^3$ .

Particulate matter (PM) can be divided in two size groups: coarse particles and fine particles. Fine particles are defined to be smaller than 2.5  $\mu\text{m}$  in aerodynamic diameter and are labelled as PM2.5. Coarse particles are defined to be smaller than 10  $\mu\text{m}$  and larger than 2.5  $\mu\text{m}$  in aerodynamic diameter and are labelled as PM10. The classification of particulate matter by taking into account their aerodynamic properties governs the transport in air and the deposition in the respiratory system and will be explained more in depth in section "Health effects from aerosols".[1]

Both the PM10 and PM2.5 fractions cause adverse health effects at elevated concentrations due to their aerodynamic properties that allow the particles to penetrate into the respiratory system and blood unfiltered, where the particles for example increase cardiopulmonary and lung cancer mortality.[1]

#### 3.1.2 Sources and size distribution

The size range of aerosol particles covers multiple orders of magnitude and is governed by the source of the emissions and processes shaping the particles. There are two types of aerosols based on their sources. Natural aerosols are emitted by sources directly correlated with different processes in nature. Anthropogenic aerosols are related to human sources, emissions correlated to human activities. Aerosol particles that are directly emitted to the atmosphere are classified as primary aerosols which means that their formation process occurs directly at the source. Aerosol particles that are formed by gas to particle conversion in the atmosphere are classified as secondary aerosols.

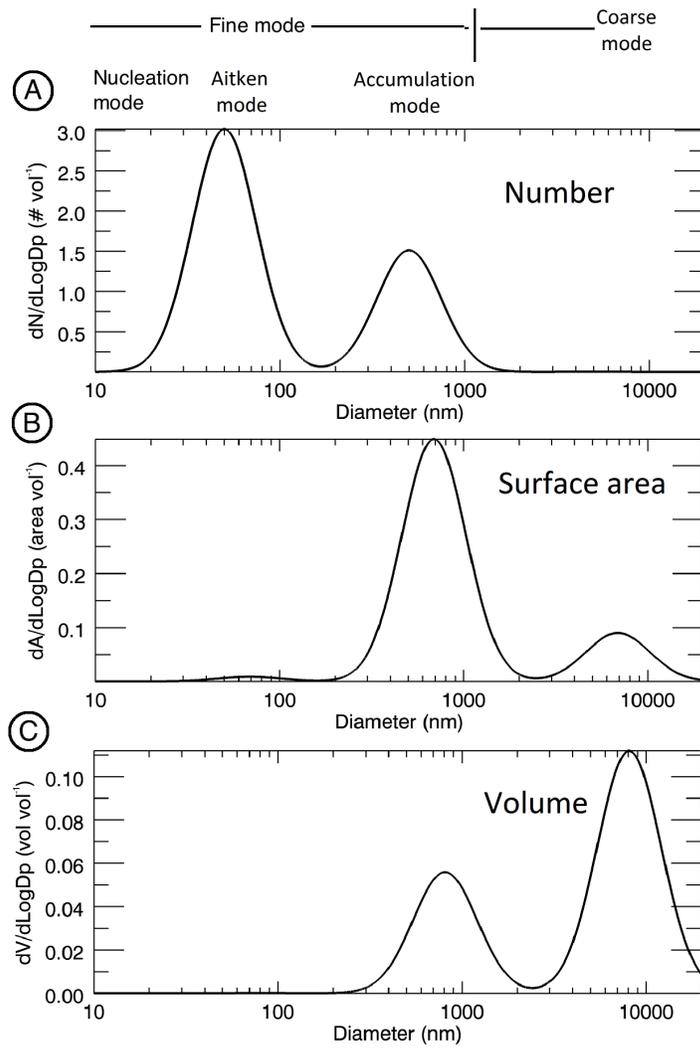
Natural primary aerosol particles originate for example from breaking up larger solid particles mechanically into wind blown dust, by the evaporation of sea spray into sea salt or by emissions from plants in the form of pollen and other plant fragments. Natural primary aerosols are coarse particles and part of the Coarse mode. The lowest size of the Coarse

mode is limited by the increasing energy needed to break up the particles into smaller sizes as their size decreases. The particles in the Coarse mode are fewer in number compared to the Fine mode and to express the aerosol particle concentration of the Coarse mode the volume concentration is therefore used. The Coarse mode dominates the volume concentrations depict in Figure 2C.[6]

Natural secondary aerosols form through gas-to-particle conversion and are often composed of sulphates and nitrates. Photochemical reactions that occur in the atmosphere between gases produces small particles that add to both the Nucleation mode and the Aitken. The smallest formed particles are nuclei and work as seed particles. These nuclei are part of the Nucleation mode together with anthropogenic primary particles which are formed when vaporised substances with low vapour pressures condensate. The primary particles in the Nucleation mode are for example created during combustion, when heavy metals are vaporised and can form nuclei. Processes like coagulation and condensation will increase the particle sizes of the nuclei and add to the Aitken- and Accumulation modes. The Accumulation mode also includes combustion particles and secondary smog particles formed in the atmosphere. The Nucleation-, Aitken- and Accumulation modes are dominating the number concentrations depict in Figure 2A and constitute together the Fine mode. Examples on other sources of anthropogenic primary aerosols that produce fine particles are industrial processes, traffic and agricultural activities. The surface area concentrations are dominated by the Accumulation mode depict in Figure 2B.[6]

The hypothetical distribution of atmospheric aerosols presented in in Figure 2 uses a logarithmic normalised particle size distribution ( $dN/d\log d_p$ ) to deal with the large span of number and size range that exist.

Natural airborne particles tend to constitute about 90 percent of the total global aerosol mass. The other 10 percent of airborne particles come from anthropogenic sources, such as emissions from traffic, industries and urban areas.[5]



**Figure 2:** The log-normal aerosol size distribution expressed by number, surface area and volume versus the particle aerodynamic diameter.

**Table 1:** Environmental quality standards in Sweden. [14]

		Daily average	Annual average
PM10	( $\mu\text{g}/\text{m}^3$ )	50 <sup>4</sup>	40
PM2,5	( $\mu\text{g}/\text{m}^3$ )	-	25
Pb	( $\text{ng}/\text{m}^3$ )	-	500 <sup>5</sup>
As	( $\text{ng}/\text{m}^3$ )	-	6 <sup>5</sup>
Ni	( $\text{ng}/\text{m}^3$ )	-	20 <sup>5</sup>
Cd	( $\text{ng}/\text{m}^3$ )	-	5 <sup>5</sup>

### 3.1.3 Processes

The previous presented size distribution of atmospheric aerosols are continuously altered by different processes affecting aerosol particles. The first process is the formation of the particles which already has been described, the gas-to-particle conversion and the photochemical reactions. Secondly there is growth which is achieved by processes as coagulation, condensation and evaporation. The collision of aerosol particles due to random motions and where a coalescence of particles is formed is called coagulation. Usually a large particle with low Brownian motion collides with a smaller particle with high Brownian motion. The coalescence that forms barely changes the form and mass of the larger particle but changes the amount of particles by removing the smaller one. Condensation makes gas or vapour molecules stick to the surface of existing particles leading to a size increase. The opposite of condensation is evaporation, which decreases particles sizes by making molecules leave the particles. The upper limit of the Fine mode is determined by the fact that coagulation is most effective for large particle numbers and condensation for large surface areas, which both decrease for large particle sizes.

The processes affecting the removal of the aerosol particles are wet deposition and dry deposition. Airborne particles in the Aitken and Accumulation mode have long residence times and transport distances in air. The particles are removed from the air by wet deposition in the form of rain, snow and hail. The Coarse mode particles are mostly deposited by dry deposition, where no precipitation is involved. This includes sedimentation and impaction.[6]

### 3.1.4 Environmental quality standards

Environmental quality standards (EQS) are set to protect human health and the environment. The EQS for outdoor air in Sweden are stated in the air quality regulation (2010:477).[13]

The air quality regulation contains limit values, target values, assessment thresholds for outdoor air in Sweden and are used as a base for EU regulations. The municipalities execute the necessary controls and if an EQS is exceeded, an action programme is started.

The environmental quality standards that are of importance for examining the metal content of airborne particles are presented in Table 1. The table presents PM limit values and four regulated heavy metals with their annual averages.

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<sup>4</sup>May be exceeded 35 times a year

<sup>5</sup>Only valid for the sampling and measuring of PM10.[14]

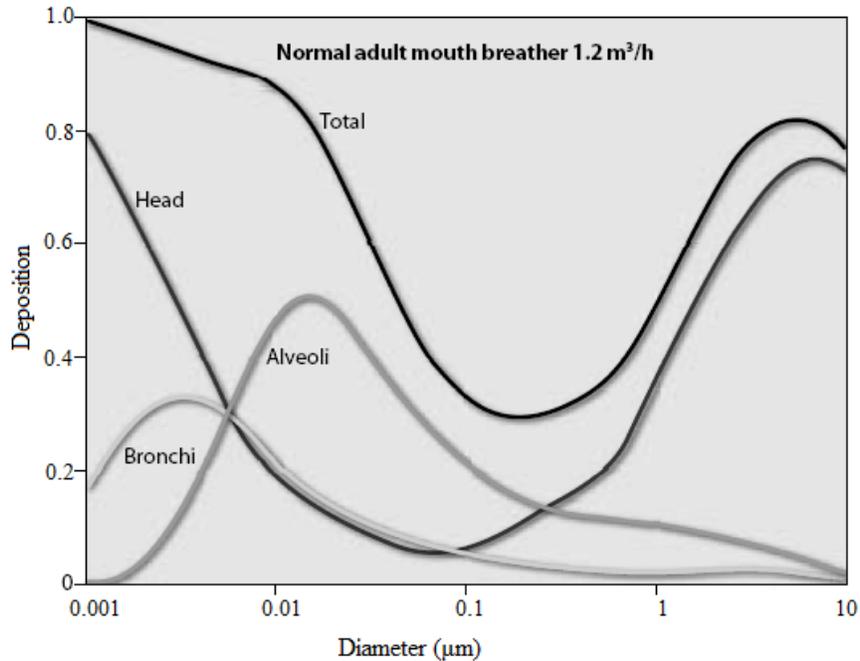
### 3.2 Health effects from aerosols

The health effects caused by high amounts of aerosol particles has been a well known problem. For example during the great smog of London in 1952, the whole city was covered in a dense smog, containing high amounts of air pollution, mostly produced by coal burning, causing an estimated death count of around 12 000 humans.[15] With an increase in knowledge from numerous epidemiological studies, the importance in measuring the fine air particulate matter became clear when correlations between mortality and lower particle loads were established. This is substantiated by estimations of about 432 000 deaths a year in Europe caused by exposure to PM<sub>2.5</sub>. [16]

As different studies demonstrate toxicological effects caused by aerosol particles, the main mechanisms and processes behind these responses are still fairly unresolved. Potential factors that cause the aerosol particles to induce adverse health effects are in the form of physical characteristics, where it shows that fine particles (PM<sub>2.5</sub>) cause a higher mortality than coarse particles, and in the form of chemical characteristics where metal content, specific surfaces and the presence of organic compounds may lead to oxidative stresses and different immune effects. [1]

The inhalation of aerosol particles is the main way of exposure to heavy metals and causes severe health effects. As the heavy metals are part of the aerosol particles, studying the deposition of particles in the lungs during inhalation is of primary concern. The deposition rates of aerosol particles are heavily dependent on particle size and particles with hygroscopic properties will take up water from the airways and lungs and grow in size. The respiratory system is divided into the head airways, where deposition often leads to the removal of particles through the digestion system, the tracheobronchial part, where the defence against deposited particles are white blood cells, and the alveolar part, where the gas exchange takes place and particles can enter the blood stream. The particles that are sufficiently small to penetrate the alveolar membranes and enter the blood are called ultrafine particles and can even be transported along nerves into the brain. The contribution in mass from the ultrafine particles is relatively small compared to PM<sub>10</sub> but due to a high surface area and large numbers, ultrafine particles are a main concern from a health aspect.[17]

In figure 3 a schematic overview of where certain sized inhaled particles deposit in the respiratory tract is presented. A great amount of particles do deposit in the head airways and are thereby removed through the digestion system to never really affect the body, while particularly PM<sub>2.5</sub> particles are able to reach deep down into the lungs and deposit in the alveolar region where the transfer to the blood stream occurs.[18]

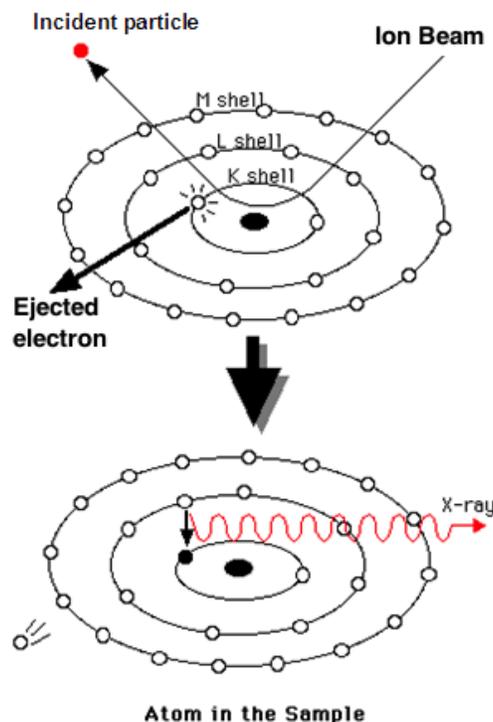


**Figure 3:** The deposition probability of inhaled particles in the respiratory system as a function of particle diameter.[18]

Certain metals, including copper, nickel and zinc, are essential elements required by humans for different biochemical and physiological functions. Different heavy metals constitute important parts in key enzymes and are needed in oxidation-reduction reactions. Insufficient intake of these metals could lead to a variety of diseases. Though on the other hand, many of these essential metals cause mayor health effects in excess amounts. Lead is classified as a non-essential metal and due to its high degree of toxicity it can lead to organ damage and is seen as a public health risk. Lead has the ability to mimic certain actions which normally are executed by calcium. This makes it able for lead to interact with proteins and cause blood, central nervous system and kidney problems with its toxicity when chronic exposure is involved. High lead exposures have reproductive effects for adults while children experience growth retardation and a lowered intelligence.[19]

### 3.3 Particle induced X-ray emission

Particle induced X-ray emission (PIXE) is a method that uses a beam of protons to excite inner shell electrons in the target atoms to generate X-ray emission characteristic of the elements. The way the method works is shown in Figure 4. First, the excitation of the inner shell electrons creates vacancies which are filled by outer shell electrons and this simultaneously creates X-rays. As the X-rays correspond to the difference in energy between the outer and inner shell, and these energies are characteristic for each element, PIXE is an elemental analysis technique. The yield of the created characteristic X-rays are proportional to the concentration of the specific element in the sample. The proton beam used in PIXE is non-destructive towards the samples and can be used on solids, liquids and aerosol filters. PIXE elemental analysis works for inorganic elements from Sodium to Uranium in the Periodic table. The characteristic X-rays for the 35 elements that can be analysed are presented in a single spectrum with a sensitivity of around one part per million. Although special factors such as peak overlap and detector efficiency affect the sensitivity, the peak to noise ratio for PIXE is better than that of electron based X-ray techniques. The limitations in elemental analyses are due to absorptions of X-rays in the detector window. The benefit of using special X-ray filters when performing the PIXE analysis is to prevent the X-ray detector from reaching too high count rates. Elements in large amounts can occupy the counting capability of the detector with their X-ray signals and cause shadowing of X-rays from minor elements. By filtering out dominant elements at lower X-ray energies in the filters, the beam current can be increased to achieve a greater element sensitivity.



**Figure 4:** Schematic overview of the characteristic X-ray creation in PIXE.

## 4 Research Methodology

### 4.1 Aerosol sampling

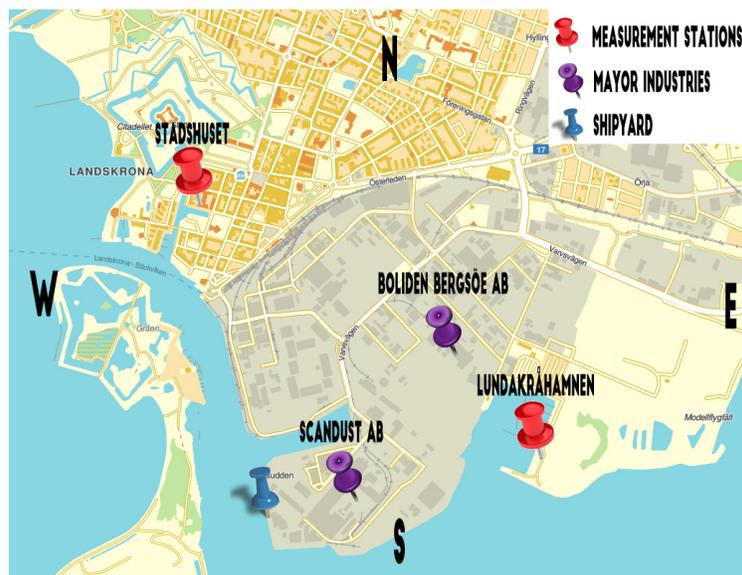
#### 4.1.1 Measurement locations

Both measurement locations are in Landskrona town on the west coast of Southern Sweden. Across the sea to the south west, about 25 km from Landskrona lies the capital of Denmark, Copenhagen, with around 2 million inhabitants in its metropolitan area. Malmö and Helsingborg, both mayor industry and harbour cities are located 30 km to the south and 20 km to the north of Landskrona respectively. To the east of Landskrona there is a major motorway that connects Malmö with Helsingborg. The measurement locations in Landskrona are shown in Figure 5.

The reference town measurements were executed at the top floor of the city hall (Stadshuset). These represent so called urban background measurements. The measurement spot at Stadshuset is presented in Figure 6. The setup was placed on a large balcony at the south side of the building. A difference from previous years is that a construction site is located in front of Stadshuset, roughly 50 m away, possibly affecting the sampled aerosols.

The second measurement location was at Lundåkrahamnen which is a small boat harbour. The measurement spot is depicted in Figure 7). The small boat harbour does not house a lot of activity during the measurement period apart from a couple of boats being painted. The industry area in Lundåkrahamnen houses two heavy metal source industries, Boliden Bergsöe AB and BEFESA Scandust AB.

The measurement locations are the same as during the previous experiments performed during 1977, 1988, 2003 and 2008.



**Figure 5:** A map of Landskrona showing the location of both measuring stations, the location of two important heavy metal source industries, the shipyard and the cardinal directions.



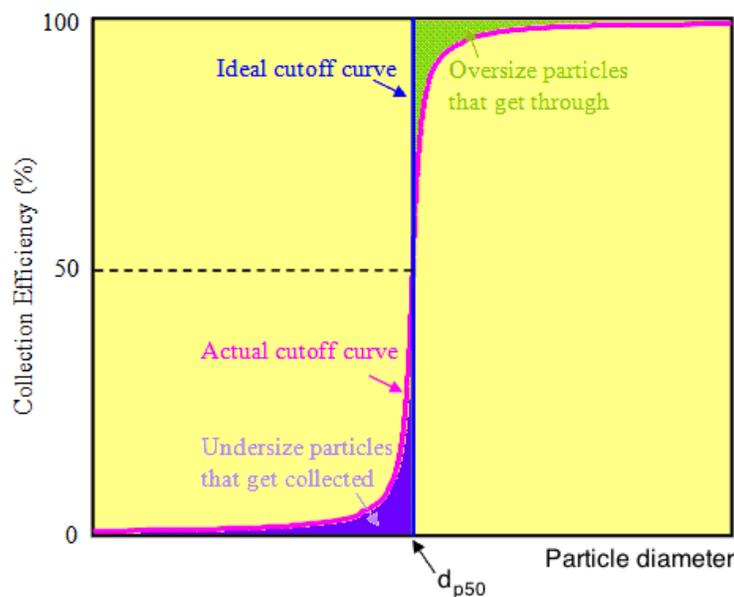
**Figure 6:** An image of the measurement setup located at Stadshuset.



**Figure 7:** An image of the measurement setup located at Lundåkrahamnen with one of the possible sources to the left in the background, Boliden Bergsöe AB.

### 4.1.2 Experimental measurement setup

The aerosol sampling was performed over a time span from 15 February to 23 March. A schematic sketch of the experimental setup at Stadshuset and at Lundåkrahamnen are depicted in Figure 9 and 10. In both experimental setups the air is pumped through a Gent sampler which creates a PM10 cutoff. This is achieved with a pre-impaction plate that removes particles larger than  $10\ \mu\text{m}$  with an efficiency of 50% at an airflow of  $16.7\ \text{l/min}$ . [20] Calculating the impactor cutoff for different airflows is of importance for this project. An impactor removes particles larger than a certain size from the airflow by mean of impaction. The airflow passes through a nozzle and the impaction plate in front of the gas stream, forms an obstacle on which particles with relatively high inertia will impact. Smaller particles with lower inertia will instead pass around the plate following the airflow. Hence the impactor plate will remove larger particles at a specific cutoff size,  $d_{p50}$ . However, in reality there exist no sharp cutoff size. The nozzle size makes it possible for some oversized particles to pass through the impaction stage while there also is a chance to collect undersized particles. The real and ideal cutoff curves for impactors is depicted in Figure 8.



**Figure 8:** The real and ideal impactor cutoff curves. [21]

The cutoff size ( $d_{p50}$ ) for an impactor can be calculated with Equation 1. To calculate the cutoff size and in that way determine the PM size that is collected the particle density ( $\rho_p$ ), the flow velocity ( $U$ ), the air viscosity ( $\eta$ ) and the nozzle diameter ( $D_j$ ) are required. The equation also needs a value for  $C_c$  though as this is a function that depends on the particle size iteration is required to obtain a  $d_{p50}$  value. Finally, Stokes number  $Stk_{50}$ , is needed (Equation 2). Stokes number takes into account the collection potential of particles.

$$d_{p50} \sqrt{C_c} = \sqrt{\frac{9\eta D_j Stk_{50}}{\rho_p U}} \quad (1)$$

$$Stk = \frac{\tau U}{D_j/2} = \frac{\rho_p d_p^2 U C_c}{9\eta D_j} \quad (2)$$

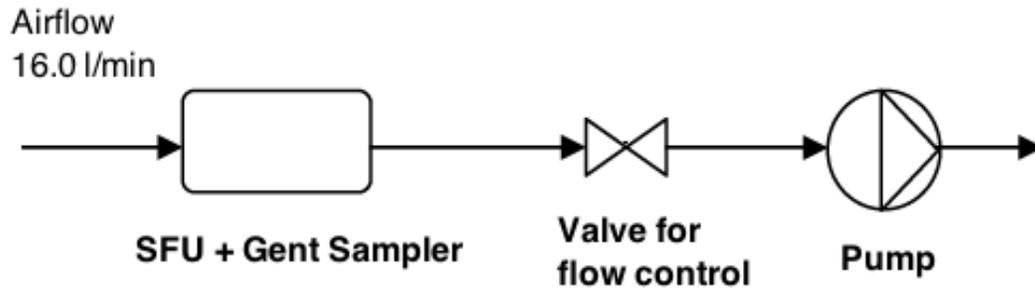
To prevent bounce-off, the plate is greased with Apiezon vacuum grease. To prevent the vacuum grease from intervening with the results yielded from the PIXE analysis it is free from Si particles. Before installing the Gent samplers, the containers were cleaned with water and ethanol to remove any contamination.

Inside the Gent samplers there are Stacked Filter Units (SFU) (Figure 11). The filter units contain two Nuclepore polycarbonate membrane filters, one with a pore size of 8  $\mu\text{m}$  and the other one with a pore size of 0.4  $\mu\text{m}$ . The first 8  $\mu\text{m}$  filter collects particles larger than 2.2  $\mu\text{m}$  diameter at an airflow of 16.7 l/min. Hence on this filter, particles between 2.2  $\mu\text{m}$  and 10  $\mu\text{m}$  diameter, in other words, the coarse fraction is collected. The second filter collects the remaining particles, in other words, the PM2.2 fine fraction. The mounting of the stacked filter units should be executed in preferably a dust-free room. As most of the filter mounting was executed at Stadshuset, no such room was present. For the mounting, a table in a less busy room was used. The stacked filter holders were opened and closed with help of special tools and these also ensured air tightness. Filter handling was always executed with tweezers. The SFU was placed in the Gent sampler and a tube was connected to the top. The Gent sampler was made water proof from the top by using a plastic bottle rain cover.

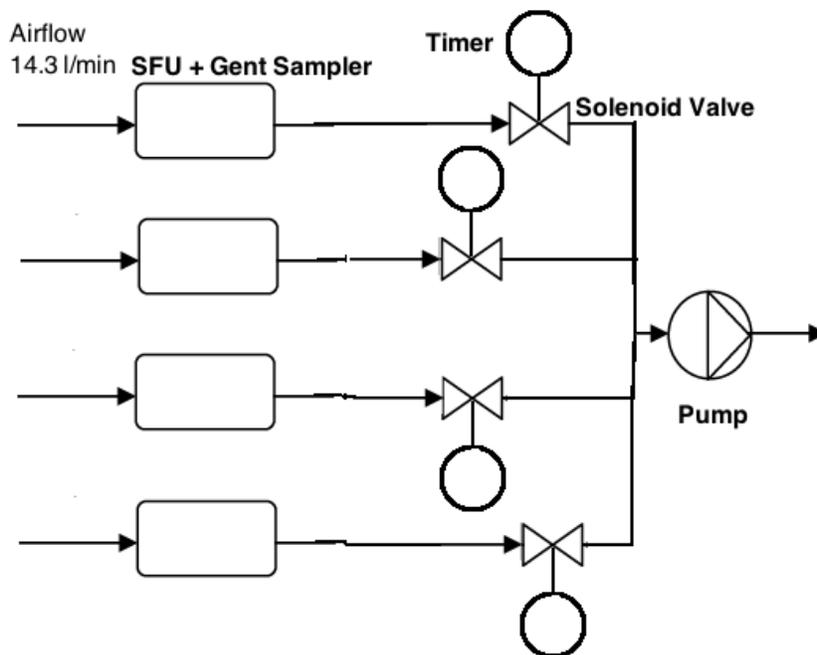
The setup at Stadshuset presented in Figure 9 also contains a valve for airflow regulation and a pump to generate the airflow. The filters in the SFU at Stadshuset are manually changed every day. When changing the filters with tweezers, they are placed in separate Petri dishes and covered with aluminium foil to prevent chemical reactions from happening due to sun light.

The measuring setup at Lundåkrahamnen presented in Figure 9 contains four SFU-units that shift automatically every day and are changed once every four days. The shifting between the different SFU-units is achieved by timers, one for each solenoid valve, which opens the valve for 24 hours (10:00 - 10:00) every fourth day.

The average airflow through the filters was about 16.0 l/min at Stadshuset. It was not possible to maintain an airflow of 16.7 l/min due to pressure drop over the filters, which increased with time due to accumulation of particles on the filters. At Lundåkrahamnen, the valves were equipped with orifices, which reduced the airflow on average to 14.3 l/min. Hence the PM10/PM2.2 collection efficiencies could not be maintained throughout the filter sampling. The flows through the filters were measured a few times during the campaign, both at the start and the end of the filter sampling.



**Figure 9:** A schematic sketch of the setup used at Stadshuset containing the SFU + Gent sampler, a valve for flow control and the pump.



**Figure 10:** A schematic sketch of the setup used at Lundåkrahamnen containing the SFU + Gent sampler, the solenoid valve which is regulated by a timer and the pump.



**Figure 11:** An image of a stacked filter unit (SFU).

The orifices in the solenoid valves were critical. To calculate the highest possible flow theoretically through a critical orifice Equation 3 is used.  $Q$  is the air flow in l/min,  $r$  is the radius of the orifice, the number 0.62 is a constant for critical flow and the number 11.7 is a constant for airflows below 20 l/min.

$$Q = r^2 * \pi * 0.62 * 11.7 \quad (3)$$

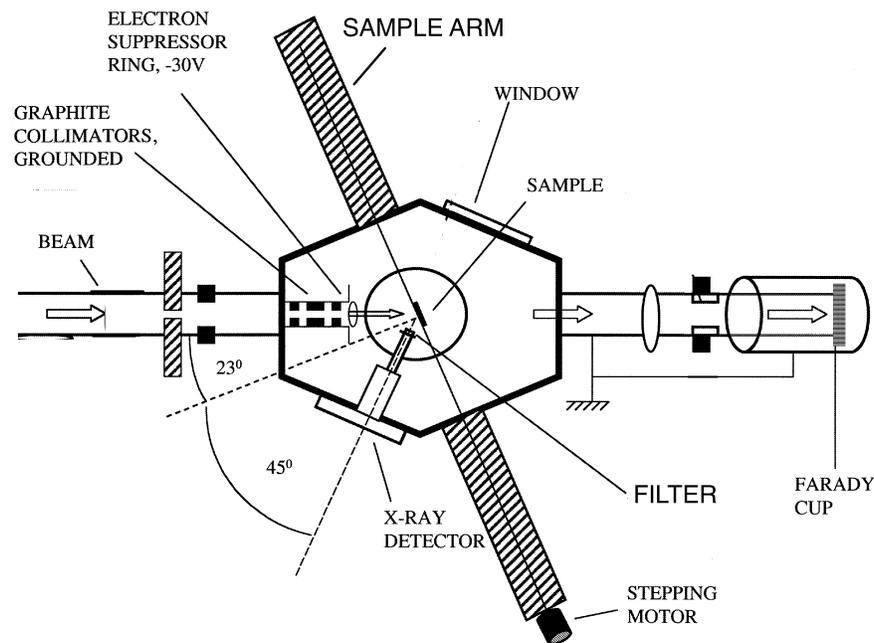
## 4.2 PIXE analysis

Before executing the PIXE analysis the samples were prepared. The filters were cut to fit the glass plates which work as the sample holders. The filters were attached to the sample holders with normal tape. The deposited areas on the filters were attached out from the holder over a circular hole in the holder. The remaining parts of the filters were kept safe in Petri dishes in case the first analysis failed. To make the PIXE analysis go smoother and prevent unnecessary correction to the equipment during executions, the samples were arranged with the same filter pore sizes in one batch. To the batches some blank filters were added. The blank filters had been mounted out in the field but never been sampled. When analysing the blank filters the contamination of sample handling is taken into consideration.

The PIXE system at Fysicum, Lund Institute of Technology, is partly presented in figure 12. The PIXE system consists of a collimated proton beam created from hydrogen plasma and brought up to speed by a 3 MV Tandem accelerator. The beam has an energy of around 2.55 MeV and while the protons pass through vacuum, focussing magnets are used to focus the beam while magnetic and electric field are used to align the beam and bring it to the sample. Cone shaped graphite collimators are used to shape the beam, giving it

well defined edges. Further by having a second collimator with a larger hole diameter than the first, scattered protons are effectively removed. All to get a well defined propagation of the beam and avoid producing and detecting X-rays from different parts of the chamber. A voltage of  $-30\text{ V}$  is applied to the last collimator to suppress secondary electrons and provide a more accurate charge integration.

Multiple samples are loaded on a sample arm that can hold 26 samples at a time. This sample arm will hold an additional 3 standard samples, one fluorescent, one aluminium and one titanium sample. These samples work as operating standards to secure the correct settings and alignment of the system. As the corrections and parameters are checked and adjusted the sample arm will move and place the samples one for one in the incident ion beam. The X-ray detector is placed outside the vacuum chamber with a filter in between. The angle between the proton beam and the detector is 68 degrees. As the proton beam has a satisfying diameter, no additional movement of at sample has to executed. The angle between the proton beam and sample is 23 degrees. The X-ray detector is of the type, Si drift detector, a thin silicon plate with a radial electric field provides high count rates, good energy resolution of and simple cooling with a Peltier element. A beryllium cover filter is used to protect the detector from high energy X-rays. A mylar filter is used to reduce the incoming X-rays from lighter elements due to their presence in higher numbers. To still measure a signal from these elements a pinhole mylar filter is used, letting through less X-rays from the lighter elements. A Faraday cup located behind the samples, catches the beam and provides an integration of the beam current. The current measured determines the number of protons hitting the Faraday cup and thereby provides the charge per sample. The samples were each ran until a charge of  $15\text{ mC}$  was reached, staying below the maximum of 4000 counts per second for the detector, leading to sampling times between 5 to 10 minutes for each sample.



**Figure 12:** An overview of the PIXE system at Fysicum, Lund Institute of Technology, showing the described parts.[22]

To identify the recorded peaks in the X-ray spectrum a quantitative PIXE imaging and analysis software is used called GeoPIXE. This tool provides an interface for interactive selection of elements and identification of X-ray lines in a spectrum. By the use of unique processes, problems like element overlaps, subtracting backgrounds and detector response effects are resolved. By automatically letting the software identify and adjust mathematical functions to the spectrum, the found peaks result in area concentrations for the found elements in  $ng/cm^2$ , the GEOPIXE fitting error in  $ng/cm^2$  and the detection limit in  $ng/cm^2$  for each element. As the concentration of heavy metals in aerosols is of importance for this project, the area concentration  $C_A$  is recalculated into a volume concentration  $C_V$  in  $ng/m^3$  with the following formula:

$$C_V = \frac{(C_A - C_B) * A}{V} \quad (4)$$

in which

- $C_A$  = area concentration of a single element in  $ng/cm^2$ .
- $C_B$  = area concentration of a blank sample in  $ng/cm^2$ .
- A = exposed filter area in  $cm^2$ .
- V = total volume of air passing through the filter in  $m^3$ .

As this project includes an experimental measurement, the risk for uncertainties due to for example measurement limitations are increased. That is why these uncertainties should be propagated. To study the uncertainty of a function based on several variables, the propagation of uncertainty for each variable uncertainty is used. In experimental measurements uncertainties occur due to different limitations and should propagate to a combined function uncertainty.

When correlations between the different uncertainties can be neglected, assuming the variables to be independent, the variance formula (Equation 5) is used to calculate the propagation of the uncertainty.

$$s_f = \sqrt{\left(\frac{\partial f}{\partial x}\right)^2 * s_x^2 + \left(\frac{\partial f}{\partial y}\right)^2 * s_y^2 + \left(\frac{\partial f}{\partial z}\right)^2 * s_z^2} \quad (5)$$

In Equation 4,  $s_f$  is the standard deviation of function  $f$  while  $s_x$ ,  $s_y$  and  $s_z$  are standard deviations for different variables.[23]

To prepare the data for the Positive Matrix Factorization (PMF) analysis, new uncertainty matrices were created. First off, as the uncertainty values received from the PIXE analysis only takes the error in fitting from the GEOPIXE program into account, an error due to the inaccuracies in the calibration of the PIXE setup should be propagated. This 7 % error is propagated with the use of Equation 4. Next the equation for uncertainty propagation and the equation for calculating the volume concentration (Equation 4 & 5) are combined. The new formula, presented in Equation 6, contains both the uncertainty for a single element in a sample, the uncertainty for the blank sample and the uncertainty from measuring the volume. The received uncertainty will be an average uncertainty.

$$s_{C_V} = \sqrt{\left(\frac{A}{V}\right)^2 * s_{C_A}^2 + \left(-\frac{A}{V}\right)^2 * s_{C_B}^2 + \left(\frac{(C_B - C_A) * A}{V^2}\right)^2 * s_V^2} \quad (6)$$

### 4.3 Data and blank handling

The selection of blank samples to correct the concentrations and calculate the volume concentrations was performed by taking samples that by accident were never run.

For Lundåkrahamnen the filters from the 28th of February were chosen. Some of the elemental concentrations of the blank sample showed higher values compared to other days, especially the elements that originate from windblown dust such as Al, Si and P. When using these blank concentrations it lead to the calculation of negative volume concentrations. To adjust for this error, other sampled filters with the lowest Al, S and P concentrations values were used as blank filters.

For Stadshuset a day with overall low concentrations was chosen as blank sample. Even for this sample certain values were higher compared to other days and again for these elements the lowest concentrations were chosen. At the same time, when choosing low concentrations for the blank sample adjustment, the concentrations are compared to the detection limit. In cases where concentrations are far below the detection limit of PIXE, values closer to the detection limit were chosen as blank values and the calculated volume concentrations for individual samples that showed negative values were set to zero. The average and median concentrations were however also based on the negative values.

To calculate the volume concentrations, the sampled air volume of every day had to be estimated. As the airflow had been sampled a couple of times during the measuring period these were used to calculate an average airflow. By using the average airflow and the time between every filter change, the sampled volume for each day was established.

### 4.4 Meteorology analysis

Figure 5 shows that winds from WSW and NW come from Scandust AB and Boliden Bergsöe AB respectively. The wind directions WSW and NW correspond to 247.5° to 315° in numerical values. By using the meteorological open database provided by SMHI (Swedish Meteorological & Hydrological Institute) the wind directions and speeds can be found for the measurement period. Though, no test site is present in Landskrona and therefore observations in Malmö and Helsingborg are used as representative meteorology data of Landskrona. This data provides hourly updates on directions and speed.

As a second part of the meteorology analysis, back trajectory analysis was executed with the use of the HYSPLIT model.[24] The back trajectory analysis was used to determine the origin of the air masses reaching Landskrona at three different heights, 3000 m, 500 m and 100 m and to describe the atmospheric dispersion between these heights. The resolution for the back trajectories was chosen to be 1 hour going 72 hours back in time. Trajectories were calculated for every third hour during each day. The trajectory plots show the trajectories at the different heights along with several meteorological parameters.

The elemental mass concentrations are daily values and will therefore be compared to daily average wind directions. To investigate the dominating wind direction every day be-

tween 10:00 to 10:00, subjective classification was made. A wind direction that clearly dominated a day was set as the direction. Other days when the wind directions were less dominant and changed between different directions, a wind direction in between was chosen. For example when the wind direction shifted evenly between WSW and SSW, the wind direction chosen was SW. Though when having a steady wind direction for the first half of the day and switched for the second half, both directions were noted. For wind directions that changed around constantly accompanied with low wind speeds, the average direction was labelled as "Switching" and meant mostly no predictable winds. The meteorology data for Helsingborg and Malmö was used to locate local sources in Landskrona and the trajectories were used to localise transboundary sources.

## 4.5 Source-Receptor modelling

Source-receptor models are multivariate statistical models which uses chemical and physical characteristics of the aerosols at the source and receptor to identify and quantify source contributions to receptor concentrations. Receptor concentrations are the sum of the source composition and source contribution products according to Equation 7:

$$x_{ij} \cong \sum_{k=1}^p a_{ik} f_{kj} \quad (7)$$

where  $x_{ij}$  is the measured concentration in ambient air of pollutant  $i$  ( $i = 1, m$ ) during sampling occasion  $j$  ( $j = 1, n$ ). Variable  $p$  corresponds to the number of relevant sources ( $k = 1, p$ ) that are significant for the model. The vector  $(a_{1k}, a_{2k}, \dots, a_{mk})$ , contains the mass fractions ( $a_{ik}$  of pollutant  $i$  in source  $k$  and is called the source profile. The  $(f_{kj}$  is the mass contributed by source  $k$  during sampling occasion  $j$ .

### 4.5.1 PMF

Positive Matrix Factorization (PMF) is a multivariate factor analysis tool that takes advantage of the fact that different sources have different compositions but that these are constant in respect to the variables that are analysed while the amplitude of the measurements will vary. This leads to the possibility to identify source profiles and source contributions solely from the the original measurement data and the uncertainty data. By utilising algorithms and the uncertainty values, the PMF model is able to produce robust non-negative solutions insensitive to outliers or values close to the detection limits. The US EPA (United States Environmental Protection Agency) PMF is the model used.[25]

## 5 Result & Discussion

### 5.1 Metal concentrations in aerosols in Landskrona

During the measurement campaign in Landskrona 61 samples were run, 35 samples at Lundåkrahamnen and 26 at Stadshuset. Less samples were run at Stadshuset because no filters were changed during the weekends. Due to disruptions in the measurement setup at Lundåkrahamnen, certain samples were never sampled and are either removed or used as blanks. In total 101 coarse and fine filters were analysed by PIXE. Based on previous executed aerosol measurements, the elements normally present in atmospheric airborne particles that have been identified by GEOPIXE are: Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Pd, Cd, Sn, Ba och Pb.[6]

#### 5.1.1 Sampling of fine and coarse particles

The previous executed measurements from earlier years both used the same sampling methodology and analysing technique which makes the results comparable. The only difference from previous campaigns is that the airflow is lower. To investigate how a lower airflow affects the results, the theoretical airflow through the valves was studied.

The theoretically achievable airflow through the solenoid valves at Lundåkrahamnen can be calculated with Equation 3 in which the orifice is set to be critical. With an orifice of 1.6 mm, the airflow (Q) through the Gent sampler is 14.60 l/min. A larger orifice would have been required to achieve a critical airflow through the solenoid valves of 16.7 l/min which was desired for the experiment.

As the flow through the filters changed over time, the flow rates were collected multiple times during the sampling period, both when inserting a new SFU and after running it for a day. The measured airflows were used to calculate average airflows and these are presented in Table 2.

With airflows lower than previous measurement campaigns a new cutoff size had to be calculated. With the use of Equation 1, a jet diameter of 10 mm and the flow rates in Table 2, new cutoff sizes were achieved which are presented in Table 3. As can be seen in Table 3, the particle cutoff at Lundåkrahamnen was roughly PM11.5 and at Stadshuset PM10.7.

With the new cutoff size calculated, it is also of importance to take a look at the effective interception diameter of the membrane filters with the changed airflow. In the following report, "Filtration of Ultrafine Chain Aggregate Aerosols by Nuclepore Filters" [26] different airflows were tested through a Nuclepore filter with a pore size of 8  $\mu\text{m}$  and ultrafine aerosols. From the report it can be concluded that an airflow of 8 l/min gives rise to an

**Table 2:** The average measured flow rates at Lundåkrahamnen and Stadshuset through the stacked filter units.

Flow rate (l/min)	Lundåkrahamnen	Stadshuset
Start	14.22	16.08
End	13.44	15.91
Average	13.83	16.00

**Table 3:** The cutoff sizes due to the pre-impaction plate and different airflows.

Flow rate (l/min)	Cutoff size ( $\mu\text{m}$ )
14.22	11.30
13.44	11.63
13.83	11.46
16.08	10.62
15.91	10.68
16.00	10.65

effective interception diameter of 1.94  $\mu\text{m}$  and that an airflow of 16.7 l/min has an effective interception diameter of 1.70  $\mu\text{m}$ . As the airflows used in this project lie between 8 l/min and 16.7 l/min, and more towards the 16.7 l/min, the effective interception diameter is barely affected in comparison to previous years measurements, and in some way, it is affected in a positive manner because actually one wants to intercept diameters from 2.5  $\mu\text{m}$  to 10  $\mu\text{m}$ .

In another report, presented by Hopke [20], the effective interception diameter achieved with an airflow of 16.7 l/min and a Nuclepore filter with a pore size of 8  $\mu\text{m}$  is reported as 2.2  $\mu\text{m}$ . This would suggest that with the current airflows used in this project the effective interception diameter of the filters would come close to the desired 2.5  $\mu\text{m}$ .

It can be concluded that the coarse particles can be classified as roughly PM11 and the fine particles as PM2. This could result in higher concentrations of certain elements in for example the fine particles when they in previous campaigns fell outside the fine particles and were classified as coarse particles. The same logic can be applied to the elemental concentrations in coarse particles where the size interval also has shifted towards a slightly higher size and now includes more larger sized particles. These increases are not substantially sufficient to disrupt the possibility to compare the measured concentrations with previous campaigns.

### 5.1.2 Comparison of average concentrations during different years

In Table 4 and 5 the average concentrations measured in fine and coarse particles at Lundåkrahamnen are compared to the measured concentrations from 2008 and 2003. The standard deviation for 2017 is also presented to allow for interpretation of the significance.

The average concentration in fine particles at Lundåkrahamnen of Cr, Mn, Fe, Ni, Cu and Zn have all increased compared to the values measured in 2008. Elements like Cr, Mn and Zn have increased more than the others, with more than double the earlier measured values. When comparing the concentrations in 2017 with 2003 in Table 4 it can be seen that most elemental concentrations have been reduced drastically over the years. When comparing the average concentrations with previous campaigns certain differences can not truly be explained because the exact wind directions for those campaigns are not known.

**Table 4:** The average concentrations in fine particles, Lundåkrahamnen ( $ng/m^3$ ). The concentrations from 2008 [6] are average concentrations measured during a 5 week period in the spring and the concentrations from 2003 [11] are also average concentrations measured during 5 weeks in the spring.

Element	2017	2008	2003	Standard deviation 2017
Al	30.3	10.5	69.8	34.6
Si	96.1	25.2	287.2	112.4
S	325.8	216.2	1541.6	264.7
Cl	410.6	0.1	192.7	665.3
K	85.7	19.5	178.4	49.8
Ca	31.4	17	149.4	35.1
Ti	0.3	1.8	9.8	2.8
V	1.0	1.7	10.3	1.1
Cr	4.1	0.2	3.3	3.9
Mn	2.3	0.6	4.3	1.4
Fe	56.7	32.7	105.4	32.6
Ni	1.6	0.99	3.7	0.9
Cu	1.7	1.0	5.1	2.3
Zn	10.1	4.4	24.7	8.8
As	0.7	0.3	7.2	0.9
Se	0.2	0.1	0.7	0.2
Br	2.8	1.4	5	2
Sr	0.4	0.	2.8	0.6
Sn	4.0	0.3	47.7	4.1
Pb	17.5	10.4	81.1	50

**Table 5:** The average concentrations in coarse particles, Lundåkrahamnen ( $ng/m^3$ ). The concentrations from 2008 [6] are average concentrations measured during a 5 week period in the spring and the concentrations from 2003 [11] are also average concentrations measured during 5 weeks in the spring.

Element	2017	2008	2003	Standard deviation 2017
Al	32.6	21.3	373.7	38.4
Si	153.4	87.6	1348.3	122.6
S	232.9	38.9	385.3	265.7
Cl	1714.7	21.8	801	1854.2
K	67.9	21.2	196.2	45
Ca	125.9	66.6	560.5	122.4
Ti	2.6	4.4	32.9	4.7
V	0.4	0.2	2.5	0.8
Cr	1,0	0.7	5.3	1.7
Mn	1.3	2.3	10.7	2.3
Fe	98.7	66.1	314.5	93.7
Ni	0.7	0.2	2.3	0.6
Cu	2.1	0.7	5.7	3.2
Zn	7.5	2.7	21.6	8.3
As	0.3	0.1	10.2	0.9
Se	0.1	0.1	-	0.1
Br	4,0	0.2	3.7	4.4
Rb	0,0	0.2	1.6	0.3
Sr	1.2	0.2	2.8	0.8
Sn	3.8	0.9	113	4.4
Pb	24.4	9.6	147	89,3

The average concentration in coarse particles at Lundåkrahamnen of Cr, Fe, Ni, Cu and Zn have all increased compared to the values measured in 2008. Though the average concentration of Cr did barely increase between 2008 and 2017. The average concentration of Mn, for a matter of fact decrease from 2008 to 2017. When comparing the concentrations in 2017 with 2003 in Table 5 it can be seen that most elemental concentrations have been reduced drastically over the years although the only element standing out is Cl. The element Cl associated with elements like Br and Sr which all are related to sea spray show increased values. Increased values compared to the average concentration from 2008 for multiple of the elements associated with windblown dust like Al, Si, K and Ca could explain the increase in Fe which also is related to windblown dust particles. The average concentration of Pb has increased in 2017 compared to 2008 but is much lower than that measured in 2003. The increase in average values from 2008 to 2017 is in most cases lower than the standard deviation and can not be proven significant.

By comparing the elemental concentrations for the different measurement campaigns it can be seen that from 2003 to 2008, all average concentrations decreased while most increased from 2008 to 2017 at Lundåkrahamnen. Reasons for these changes in concentration from

one year to another could be due to different wind directions or changed wind speeds. Wind directions are important for the presence of different elements. For example winds from farmlands often lead to an increased amount of soil dust.

By studying the average concentrations further it can be discovered that the concentration of Cl in coarse particles is higher than 2003 and 2008. This would suggest an increased amount of days with wind directions directly from the ocean as the element Cl is associated with mainly sea spray. Increased wind speeds would also stimulate the production of sea spray and increase the concentration of measured Cl.

In Table 6 and 7 the average concentrations measured in fine and coarse particles at Stadshuset are presented and compared to the concentrations measured in 2008, 2003, 1988 and 1977. The standard deviations for 2017 are also presented.

Table 6 shows and increase in average concentration for most elements compared to the previous measurement campaign in 2008. The concentrations measured are however lower than most of the concentrations measured in 2003, 1988 and 1977. One element that stands out with a relative high concentration compared to all other campaigns is Cl. The high average concentration of Cl is affected by the wind directions during the measurement campaign where winds from the sea increase the Cl levels.

Elements like Al, Si, K, Ca and Ti have increased in average value compared to 2008. A reason for this can be the construction site located in front of Stadshuset. At the construction site during the measurements, ground work is being executed with big machines and this could have an impact on the amount of windblown dust being released and emitted. The construction site can be seen as an extra source of dust. The increased concentrations for the elements mentioned are all typical for soil. Though windblown dust is classified as coarse particles and should therefore mainly affect the PM10 concentrations. The elements Mn, Fe and Ni have however decreased in average concentration for fine particles at Stadshuset. As these metals have shown to be typical for a source related to Scandust AB in previous reports, it can be explained why the concentrations are lowered. During the measurement campaign, Scandust AB has been closed. During the measurement period no recycling processes have been executed at Scandust AB and the production has been on hold.[9] This production stop would mean no emissions at all, or at least lowered emissions of the associated metals.

The average PM10 concentrations at Stadshuset presented in Table 7 show only elevated values for some of the elements related to windblown dust. For example are Al and Ca elevated but are Si, K and Ti decreased in concentration. Elements like Cr, Mn, Fe and Ni have all decreased in average concentration compared to the concentration from 2008. The average concentration for Zn has barely increased from 2008 and is still at a fairly low level compared to the previous years. The average concentration of Cu has never been measured this high compared to all other years, with a value of  $14.9 \text{ ng}/\text{m}^3$ . Compared to the average concentration in 2008, this is an increase of 7 times. By comparing the measured average concentrations of Pb the coarse particles show a decreasing trend starting in 1977 with  $120 \text{ ng}/\text{m}^3$  with a value of  $4.9 \text{ ng}/\text{m}^3$  in 2017.

Even for the average values at Stadshuset the increase from 2008 to 2017 falls below the standard deviation and can not be proven of significance. As already has been mentioned multiple times are the average concentrations comparable with previous measurement campaigns but because these concentrations are highly affected by just a few samples with elevated values other statistical values could be of favour when creating a overview of the

**Table 6:** The average concentrations in fine particles, Stadshuset ( $ng/m^3$ ). The concentrations from 2008 [6] are average concentrations measured during a 5 week period in the spring and the concentrations from 2003 [11] are also average concentrations measured during 5 weeks in the spring. The concentrations in 1988 [6] are average concentrations measured during the winter and spring for 4 weeks and the concentrations from 1977 [6] are recalculated annual concentrations that were sampled over 16 days during a half year period.

Element	2017	2008	2003	1988	1977	Standard deviation 2017
Al	38.7	10.4	50.4	-	-	39.3
Si	69.5	39.9	147	-	-	136
S	396.6	277.8	1241	1500	1800	80.1
Cl	755.9	0.2	161.1	24	160	1451
K	76.0	29.9	144.3	79	150	25.6
Ca	35.8	30	102.4	47	200	125.2
Ti	3.4	2.7	7.8	4	22	6
V	1.5	2.6	9.3	3	13	0.9
Cr	3.7	2.3	1.8	4.2	3.9	2.4
Mn	1.9	2.7	2.7	3.7	33	1.9
Fe	37.9	53.8	61.9	62	400	127.9
Ni	1.2	1.8	3.3	2	6.9	0.5
Cu	2.4	1.6	8	1.8	13	55
Zn	10.4	9.2	22.4	34	100	25.3
As	0.8	0.2	3.9	-	-	0.3
Se	0.1	0.1	0.8	-	-	0.1
Br	2.1	1.4	3.8	31	21	3.4
Sr	0.6	0.1	0.4	-	-	0.8
Sn	0.3	0	35.9	-	-	4.9
Pb	4.7	4.2	38.1	38	420	9.7

**Table 7:** The average concentrations in coarse particles, Stadshuset ( $ng/m^3$ ). The concentrations from 2008 [6] are average concentrations measured during a 5 week period in the spring and the concentrations from 2003 [11] are also average concentrations measured during 5 weeks in the spring. The concentrations in 1988 [6] are average concentrations measured during the winter and spring for 4 weeks and the concentrations from 1977 [6] are recalculated annual concentrations that were sampled over 16 days during a half year period.

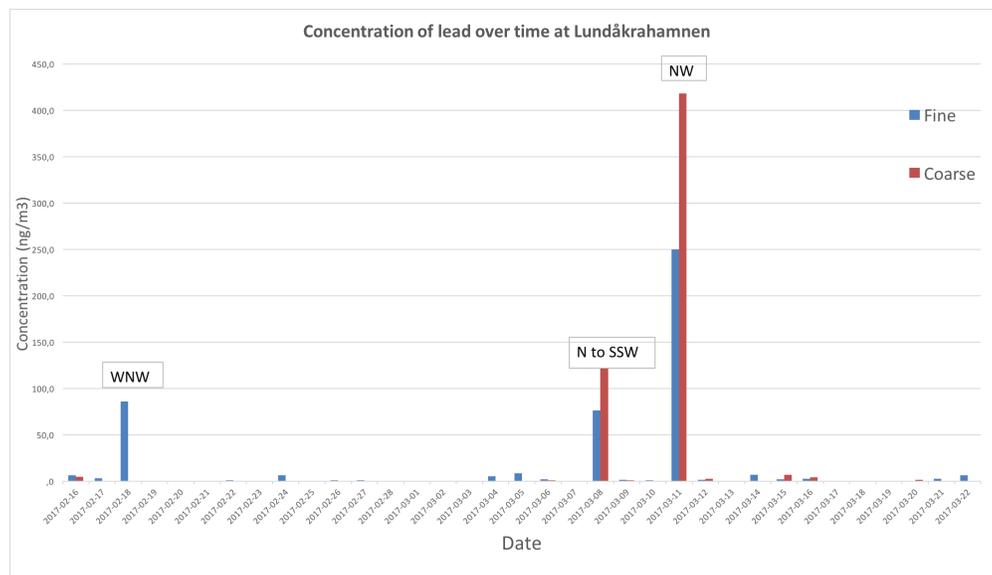
Element	2017	2008	2003	1988	1977	Standard deviation 2017
Al	67.4	49.8	217.2	139	-	29.2
Si	189.0	192.7	843.2	529	-	47.3
S	146.8	39.4	225.8	278	200	450.9
Cl	1787.5	38.6	960.1	259	340	887.7
K	43.7	46.7	147.8	139	120	62.4
Ca	145.1	125.6	512.7	338	460	22.1
Ti	7.8	9.6	30.1	22	46	3.4
V	0.4	0.4	1.8	-	3.6	1.2
Cr	3.6	4.3	1.4	3.8	1.7	2.9
Mn	1.6	6.2	7.1	7.7	8	1.5
Fe	103.9	149.2	193.6	241	400	37.6
Ni	0.7	1.2	0.9	1.2	2.3	0.5
Cu	14.9	2.2	6	3	11	5.9
Zn	11.7	11.2	15.7	18	39	12.8
As	0.1	0.2	5.6	-	-	1.3
Se	0.1	0.2	0.6	-	-	0.2
Br	2.8	0.4	2.4	2.9	5	1.8
Rb	0	0.5	1.1	-	-	0.4
Sr	1.0	0.6	1.8	-	-	0.5
Zr	0.4	0.2	0.7	-	-	0.3
Sn	4.4	0.5	98.8	-	-	3
Pb	4.9	7	44.4	25	120	8.7

correct aerosol situation in Landskrona. Therefore are other important statistical values such as Median, 10th Percentile, 90th Percentile, Max and Min presented in Table 13 to 16 in Appendix A. Most median values are lower than the average value due to the few samples with elevated concentrations and more samples with lower concentrations. Many Min values are zero which is due to the fact that the concentrations are simply too low to be measured with PIXE and end up far below the detection limit. That means that after correction with the chosen blank samples, the negative values were set to zero.

In Table 13 and 14 in Appendix A, the statistical concentrations for Lundåkrahamnen are presented. The 90th Percentile for Pb in coarse particles is even lower than the average concentration. This truly proves that the concentrations on certain days, when sources and wind directions align, are very high. Though if one looks at the Max measured Pb concentration in coarse particles at Lundåkrahamnen is it still below the allowed annual average concentration limit set by the authorities in the environmental quality standards. The 90th Percentile for the concentration of Pb in fine particles is higher than the average concentration and shows that the emissions of Pb occur at least during 10 % of the measured days but possibly more when the wind directions are in other directions.

### 5.1.3 Metal concentrations and wind analysis

When comparing the different average concentrations for several metals it is important to know that these are highly affected by a few samples with increased values. These increased values have multiple reasons. Wind directions play a crucial part as different local aerosol sources are based at different locations in Landskrona. When the wind direction corresponds to the direction of one of the major industries elevated values are recorded. For example when looking at the concentrations of element Pb in Figure 13 at Lundåkrahamnen, it can be seen that elevated values are present in both the fine and coarse particles on 8 March and 11 March. In the fine particles there is even an elevated concentration on 18 February. By examining the different wind directions for these dates from Table 8 it can be examined that high Pb concentrations correspond to wind directions between N and W and mostly NW. When these wind directions are compared to the map of the area, the industry and aerosol source in the mentioned direction is with most certainty Boliden Bergsöe AB.

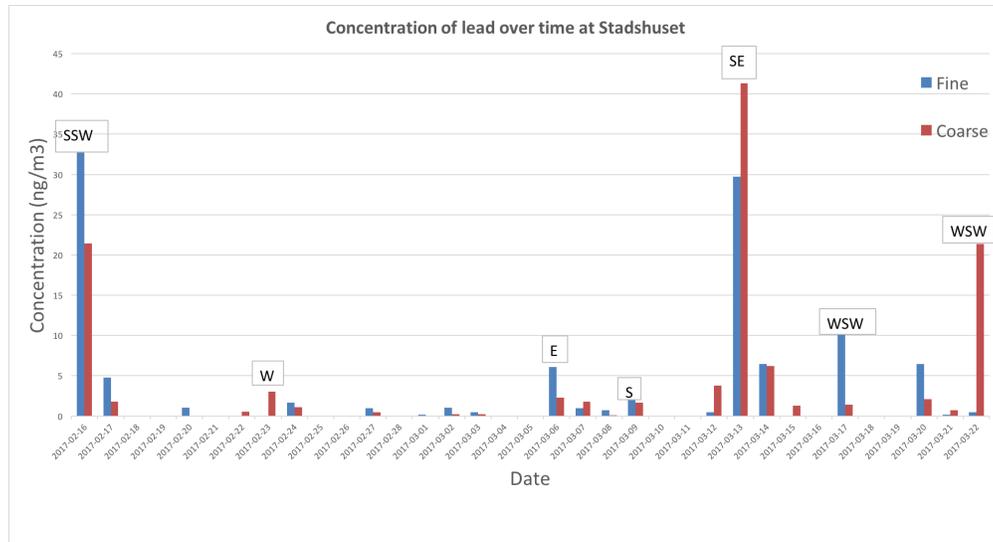


**Figure 13:** The concentration of lead at Lundåkrahamnen during the measurement period.

Boliden Bergsöe AB is placed in a different wind direction towards Stadshuset than towards Lundåkrahamnen, namely SE. Hence, elevated Pb concentrations are expected during SE winds at Stadshuset. Indeed, Figure 14 shows that this is the case during 13 March, a day with SE winds. However the same figure shows elevated Pb concentrations for winds from other directions on 16 February, 23 February, 6 March, 9 March 17-19 March and 22 March. Even though these days do not have winds from SE, both the wind from S and E could possibly have been affected by Boliden Bergsöe AB due to local winds in Landskrona, unexplained by the average wind direction from Table 8. On the other days, the elevated Pb concentrations are measured for wind directions from mostly WSW. Similar Pb concentration peaks are visible on 24 February, 5 March and 14-15 March in Figure 13 for Lundåkrahamnen when having similar wind directions.

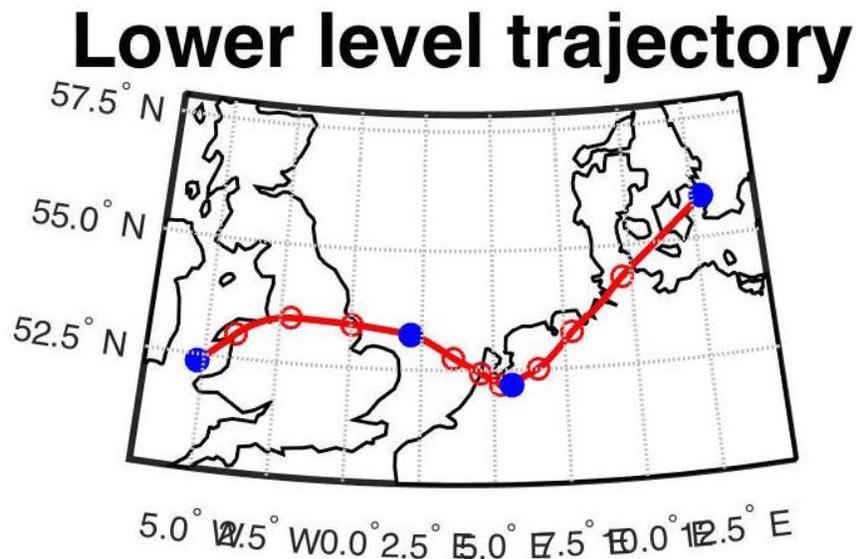
**Table 8:** The average wind directions per day composed from three different sources.

Date	Trajectory 100m	Helsingborg	Malmö
2017-02-16	SSW	S	SSW
2017-02-17	SW	SW	WSW
2017-02-18	W	WNW	WNW
2017-02-19	WSW	SW	W
2017-02-20	WSW	WSW	W
2017-02-21	W	W	W
2017-02-22	W	W to SW	W
2017-02-23	W	W	W
2017-02-24	Changing	W to S to E to NW	W to S to E to NW
2017-02-25	WNW	WNW	WNW
2017-02-26	WSW	SSW	WSW
2017-02-27	WSW	WSW to SSW	SW
2017-02-28	S	S	SSW
2017-03-01	SW	SSW	SSW
2017-03-02	SW	SSW	SW
2017-03-03	W	SW	WSW
2017-03-04	SW to SE	E	SW to SE
2017-03-05	E	E	E
2017-03-06	ENE	E	E
2017-03-07	NE	NE	NE
2017-03-08	NE to SW	N to SSW	N to SSW
2017-03-09	SSW	S	SSE to SSW
2017-03-10	W	SSW to NW	SW to WNW
2017-03-11	NW	NW to N	NNW
2017-03-12	SE	SW to SE	Changing
2017-03-13	SE	SE	SE to none
2017-03-14	SW	S to SW	S to SW
2017-03-15	W	W	W
2017-03-16	W	W	WNW
2017-03-17	WSW	SW	WSW to W
2017-03-18	WS	SW	WSW
2017-03-19	NW	NW	NW
2017-03-20	SW	SE	S to W
2017-03-21	WSW	SW	W to WSW
2017-03-22	WSW	WSW	WSW
2017-03-23	WSW	W to E	WSW to none



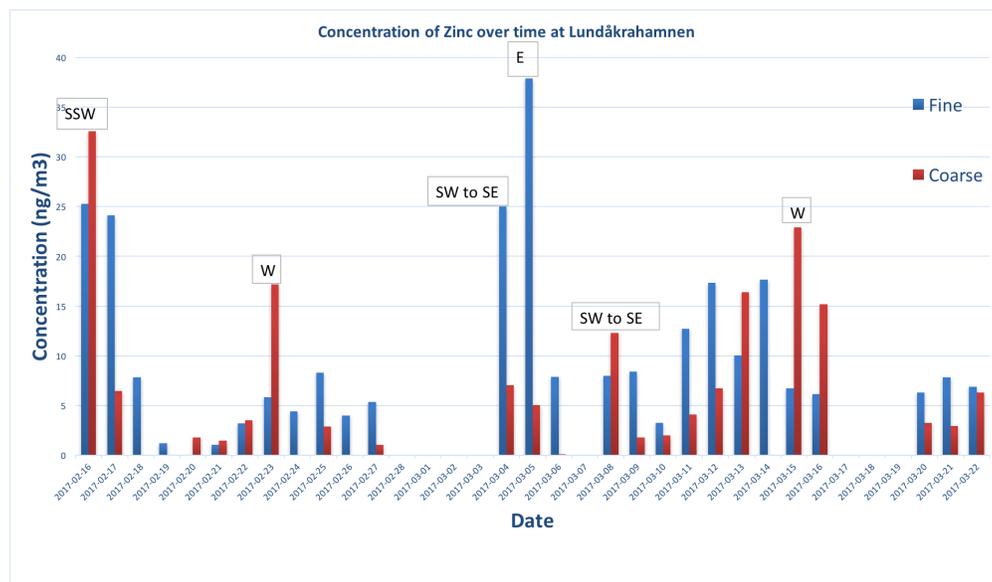
**Figure 14:** The concentration of lead at Stadshuset during the measuring period.

The Pb concentrations from WSW could be explained by a transboundary source. When studying the trajectories for these days, it can be seen that the trajectories come across both Öresund, with heavy marine traffic, and Copenhagen. One of the trajectories has been depicted in Figure 15 and corresponds to the low level trajectory on the 17 March. Similar to this trajectory, the trajectories on the other days with WSW winds all pass across Copenhagen.



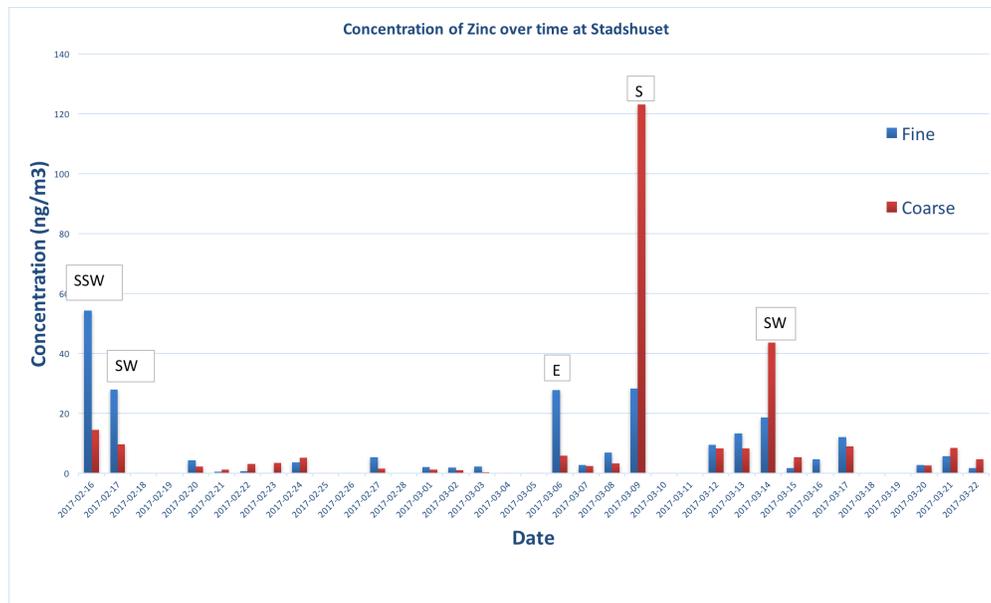
**Figure 15:** The trajectory plot for 100 m ending at Landskrona on 17 March.

In Figure 16 the concentration of Zn over time at Lundåkrahamnen is presented. The higher concentrations of Zn occur more frequent compared to high concentrations of Pb. The wind directions that correspond to high Zn concentrations lie between E and W. The highest Zn concentration was measured in fine particles for a wind direction from E on 5 March and the highest Zn concentration in coarse particles was measured for a wind direction from SSW on 16 February. When the wind direction came from W, like on 23 February and 15 and 16 March the concentration of Zn is elevated in coarse particles. The elevated concentrations from E correspond to winds from the motorway going by Landskrona. Both Zn and Cu are elements related to road traffic and could definitely affect the measurements at Lundåkrahamnen.[27] The elevated concentrations from the W could come from the shipyard which is located in this direction. The concentrations of Zn could be affected by the near vicinity of the measurement setup in Lundåkrahamnen. This is a harbour for hobby boats and in the spring most boats are painted with boat paint. Boat paint contains metals like Pb, Zn and Cu.[28] During boat repair, paint particles could become airborne and affect the measurements.



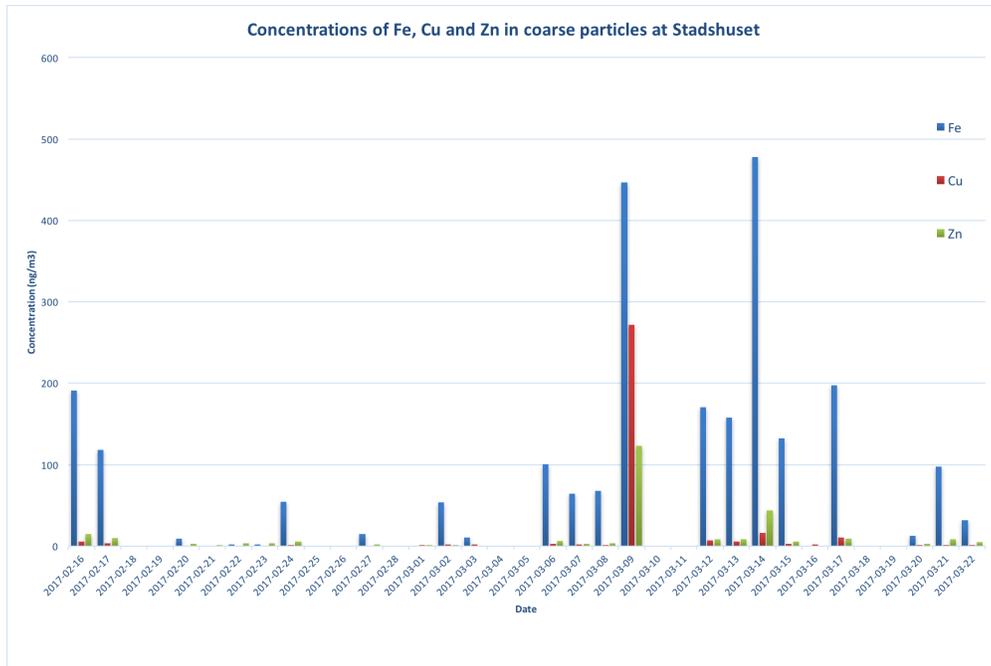
**Figure 16:** The concentration of zinc at Lundåkrahamnen during the measurement period.

In Figure 17 the concentration of Zn over time at Stadshuset is presented. Several concentration peaks were measured for Zn. The highest Zn concentration measured in coarse particles was on 9 March with a wind direction from S. The wind directions that correspond to high Zn concentrations lie between E and SW. Though by studying Figure 17 and comparing the measured concentrations with the wind directions on each day, no real correlation can be found. The low concentration that have been measured between 20 February and 3 March are dominated by W wind direction but do also contain a few days with SW wind direction. W and SW winds from Stadshuset would mean sea winds and therefore low Zn concentrations are expected. The SSW winds that show elevated levels could be affected by transboundary sources from Copenhagen which lies in this direction.

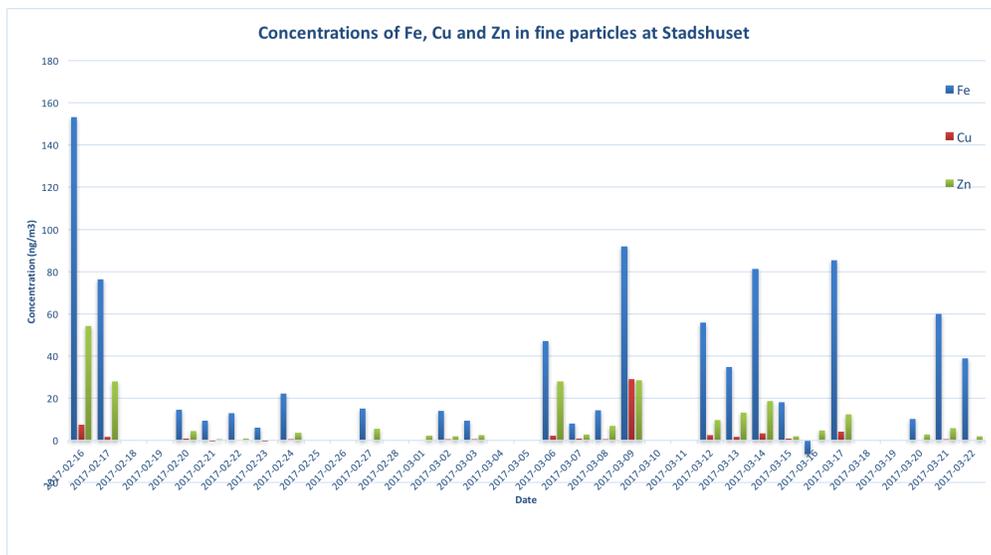


**Figure 17:** The concentration of zinc at Stadshuset during the measurement period.

The highest concentration of Zn measured on 9 March is more than double as high as the second highest Zn concentration. By comparing the Zn concentration to other elemental concentrations it was found that on 9 March a peak in concentrations occurred for both Fe, Cu and Zn at Stadshuset with a wind direction from S (Figure 18 and 19). Both in fine particles and coarse particles the three elements show a correlation between their concentrations and suggest a similar source. As the concentration for Fe, Cu and Zn are high, a local source is to be expected. Following the S wind direction in Landskrona from Stadshuset the sources that come to mind and correspond with the elements measured are Scandust AB or the shipyard. The reason for suggesting the shipyard is because Scandust AB is supposed to have been closed for the whole measurement campaign. In the report from 2008 ([6]) it was suggested that Cu concentrations could originate from the shipyard, located to the west of Scandust AB. In other ways, this peak in the concentrations of Fe, Cu and Zn is interesting and can not be fully explained. It does seem like the Zn and Cu concentrations do originate from the shipyard area by comparing the wind directions with the measured peaks for Lundåkrahamnen in Figure 16.



**Figure 18:** The concentration of iron, copper and zinc in coarse particles at Stadshuset during the measurement period.



**Figure 19:** The concentration of iron, copper and zinc in fine particles at Stadshuset during the measurement period.

## 5.2 Sources to aerosols in Landskrona

To create an overview of the number and characteristic elements of the relevant sources contributing to aerosols in Landskrona, the report from 2008 ([6]) was studied. Along with knowledge about characteristic elements and meteorology data the following discernible source types were identified: Combustion (S, V, Ni), Soil dust (Si, K, Ca, Fe), Sea spray (Cl, Br, Sr), Metal source 1 (Pb), Metal source 2 (Cu/Zn) and for fine particles long range transported pollution (LRT).[29]

PMF modelling was used to determine the qualitative contribution from the sources. For the modelling of the fine particles six sources were used and for the coarse particles five sources were used. First off the input data was analysed and the following elements were labelled as bad due to their signal-to-noise ratio which was in most cases equal to zero: Ga, Ge, Y, Zr, Pd, Cd, Sn and Ba. The concentration time series was examined but no additional samples were excluded because of unusual concentrations.

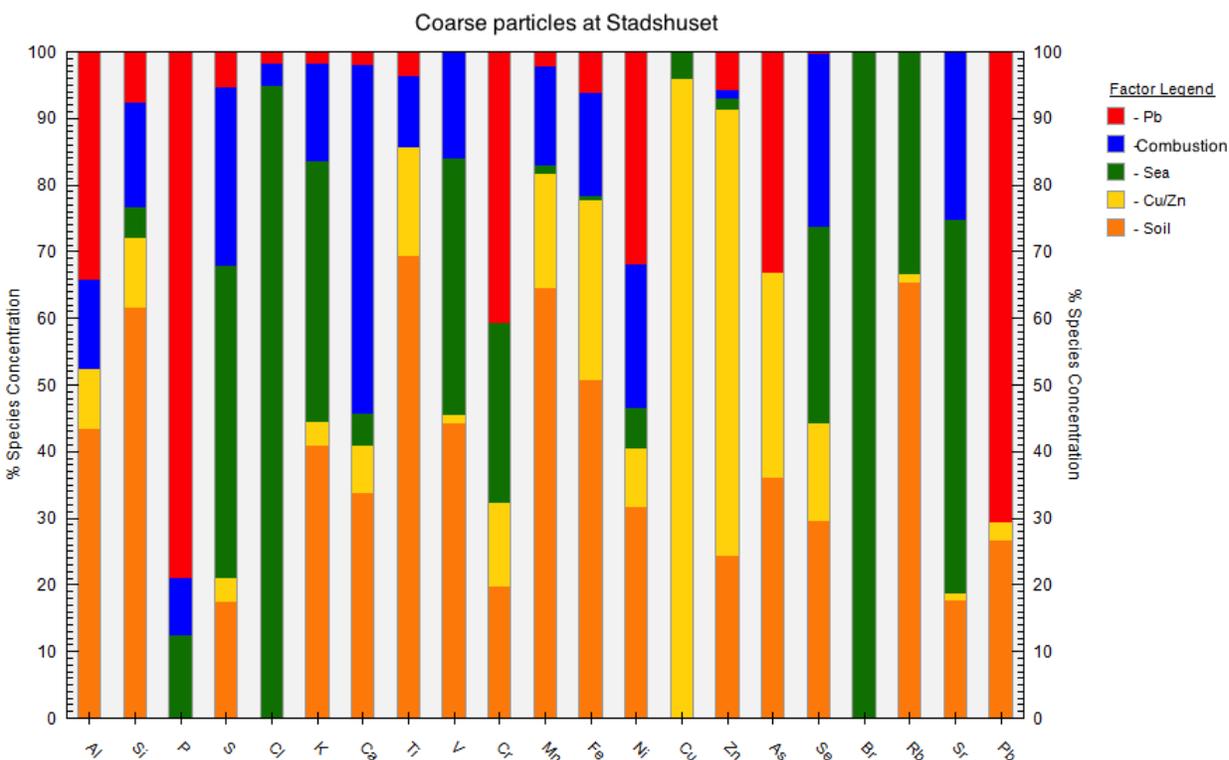
In PMF the FPEAK parameter modifies the polarisation between source profiles. By changing FPEAK between 1 and -1, where 1 is less polarisation, 0 is the standard case and -1 is increased polarisation, different source profiles can be achieved and compared. In this project the source profiles are created with FPEAK = 0.

The absolute contribution from each respective source to the concentration of an element in coarse particles at Stadshuset is presented in Table 9. The same table shows the percentage of the concentration that has been modelled compared to the average measured concentration. The PMF-model has trouble modelling the correct concentrations for Al, P, S, Cl, Ti, Cr, Se, and Sr as can be seen by having a percentage below 50. This could be the result from the few samples with elevated concentrations from local sources in Landskrona which make it harder for the PMF model to explain the time variation for these elements. For Rb the modelled concentration is higher than the average measured concentration which is zero.

**Table 9:** Source apportionment according to the PMF model of coarse particles at Stadshuset. Concentration units are ( $ng/m^3$ ) for all elements.

Element	Sources					Sum		Model/ Measured
	Pb	Combustion	Sea	Cu/Zn	Soil	Model	Measured	
Al	7.50	2.96	0.00	2.00	9.53	21.99	67.4	32.6%
Si	8.92	18.15	5.32	12.27	71.47	116.12	189.0	61.4%
P	3.14	0.34	0.49	0.00	0.00	3.98	12.9	30.8%
S	3.10	15.88	27.80	2.15	10.32	59.26	146.8	40.4%
Cl	10.97	20.71	574.15	0.00	0.00	605.83	1787.5	33.9%
K	0.52	4.05	10.84	0.97	11.35	27.73	43.7	63.5%
Ca	1.65	43.43	3.99	6.03	28.05	83.14	145.1	57.3%
Ti	0.12	0.34	0.00	0.53	2.25	3.25	7.8	41.7%
V	0.00	0.07	0.17	0.01	0.19	0.44	.4	109.2%
Cr	0.66	0.00	0.44	0.20	0.32	1.61	3.6	44.8%
Mn	0.02	0.16	0.01	0.18	0.70	1.08	1.6	67.3%
Fe	3.66	9.19	0.42	15.99	30.12	59.37	103.9	57.1%
Ni	0.13	0.09	0.02	0.04	0.13	0.40	.7	57.6%
Cu	0.00	0.00	0.41	9.55	0.00	9.96	14.9	66.9%
Zn	0.37	0.08	0.11	4.32	1.58	6.46	11.7	55.2%
As	0.03	0.00	0.00	0.02	0.03	0.08	.1	80.6%
Se	0.00	0.01	0.01	0.01	0.01	0.03	.1	34.8%
Br	0.00	0.00	1.12	0.00	0.00	1.12	2.8	40.2%
Rb	0.00	0.00	0.01	0.00	0.02	0.03	0	-
Sr	0.00	0.11	0.25	0.00	0.08	0.44	1.0	44.2%
Pb	1.81	0.00	0.00	0.07	0.68	2.56	4.9	52.3%

In Figure 20 the fingerprint for the coarse particles at Stadshuset are presented with the modelled elements and the correlated sources. It can clearly be seen that Pb is part of one source and that the second metal source contains mostly Cu and Zn. Though as the following fingerprint figure was created with a few poorly modelled elements it can be seen that the S should mainly come from a combustion source. In this case it is modelled to come from the sea. This problem occurs due to the fact that a main combustion source at Landskrona is from marine traffic. This leads to the confusion by the PMF where it merges the combustion elements and sea elements into the sea source. In some cases this problem can be solved by using a FPEAK of -1 to polarise the source profile but in this case this did not solve it.

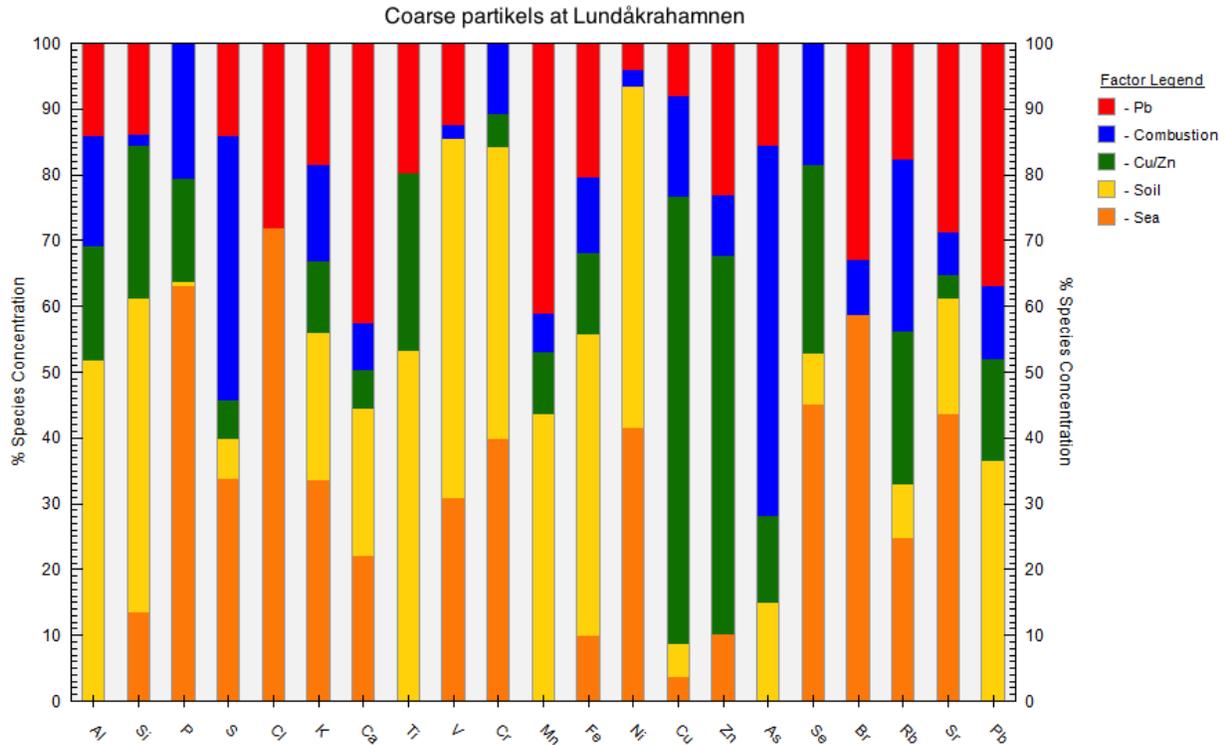


**Figure 20:** The factor fingerprint for coarse particles at Stadshuset.

When looking at Table 10 it can be seen that the PMF model had trouble modelling element Pb. This shows in the sectioning of element Pb into many different sources. By just looking at the sources and the elements they include it can be difficult to create a good overview. When Table 9 to 12 are compared it can be seen that elements that seem to be part of a source are in fact not. For example in Table 10 Mn is partly emitted by source Pb but in the other tables this correlations is not present. When comparing Figure 20 to 23 it seems like As is partly emitted by the metal source labelled as Cu/Zn. The element As is also emitted by the source labelled as combustion. Element Fe looks to be emitted by most sources but is mainly emitted by Soil dust.

**Table 10:** Source apportionment according to the PMF model of coarse particles at Lundåkrahamnen. Concentration units are ( $ng/m^3$ ) for all elements.

Element	Sources					Sum Model	Measured	Model/ Measured
	Pb	Combustion	Cu/Zn	Soil	Sea			
Al	4.87	5.81	6.02	17.97	0.00	34.67	32.61	106.3%
Si	31.35	3.75	52.71	107.82	30.32	225.94	153.42	147.3%
P	0.00	1.53	1.15	0.05	4.66	7.40	11.57	63.9%
S	39.25	110.50	15.93	16.97	93.20	275.85	232.94	118.4%
Cl	471.76	0.00	0.00	0.00	1207.70	1679.46	1714.75	97.9%
K	15.75	12.53	9.24	19.06	28.52	85.09	67.87	125.4%
Ca	65.81	11.18	9.02	34.42	34.29	154.72	125.91	122.9%
Ti	1.18	0.00	1.60	3.18	0.00	5.97	2.57	232.0%
V	0.11	0.02	0.00	0.47	0.26	0.86	0.38	225.8%
Cr	0.00	0.18	0.08	0.74	0.67	1.67	1.04	161.5%
Mn	0.93	0.13	0.21	0.98	0.00	2.25	1.32	170.9%
Fe	28.53	15.86	17.34	63.66	13.87	139.26	98.68	141.1%
Ni	0.02	0.01	0.00	0.24	0.19	0.45	0.67	67.1%
Cu	0.26	0.49	2.19	0.16	0.12	3.22	2.14	150.2%
Zn	2.19	0.88	5.47	0.00	0.96	9.50	7.51	126.6%
As	0.08	0.29	0.07	0.08	0.00	0.52	0.26	201.1%
Se	0.00	0.02	0.03	0.01	0.05	0.11	0.07	145.1%
Br	1.25	0.32	0.00	0.00	2.22	3.79	3.97	95.6%
Rb	0.03	0.04	0.04	0.01	0.04	0.17	0.04	397.1%
Sr	0.36	0.08	0.04	0.22	0.55	1.25	1.20	104.3%
Pb	0.66	0.20	0.27	0.65	0.00	1.77	24.45	7.2%



**Figure 21:** The factor fingerprint for coarse particles at Lundåkrahamnen.

For the coarse particles at both Stadshuset and Lundåkrahamnen there were 5 sources found. The sea source has characteristic elements like Cl and Sr but does also contain amounts of soil dust elements. The Pb source does mainly contain Pb concentrations but again also amounts of soil dust. The next metal source modelled was a source with mainly concentrations of Cu and Zn. This source also contains higher amounts of other heavy metals like Cr. The soil source contains characteristic soil elements like Al, Si and Fe but is also affected by amounts of V and Ni which comes from marine traffic. The final source for coarse particles in Landskrona is labelled as a combustion source but as it also contains amounts of both soil and sea elements at Stadshuset it becomes more unclear. At Stadshuset this source is probably more related to soil/sea from another long-range transport. At Lundåkrahamnen this source instead contains both soil and combustion specific elements and could come from some local source, but should anyway be labelled as combustion/soil.

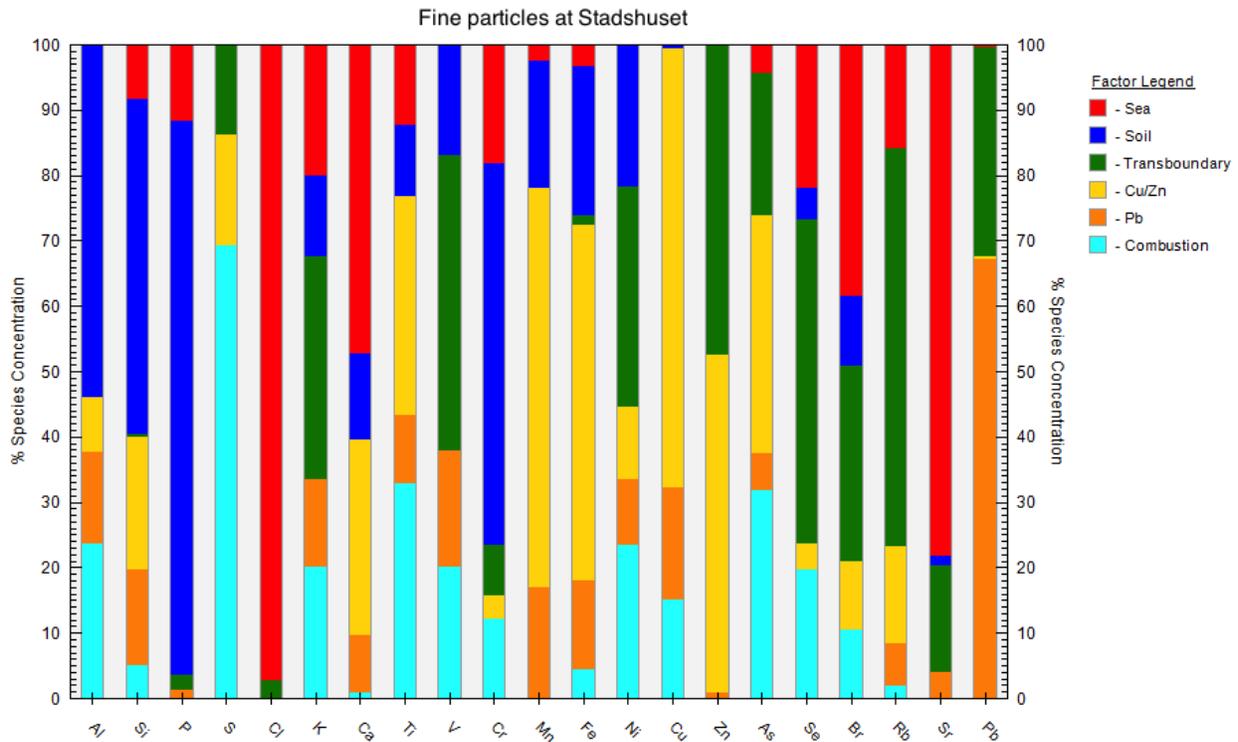
When modelling the fine particle concentrations in Landskrona, it showed that six sources gave a better model-to-measured ratio. This meant including a sixth source labelled as long range transport pollution. This source contains transboundary pollutions and affects many of the metal concentrations. The model/measured ratios are presented in Table 11 and 12. Both Figure 22 and 23 show that does this source only affects the concentrations of Pb, Cu and Zn with small amounts while the local sources have a big effect.

**Table 11:** Source apportionment according to the PMF model of fine particles at Stadshuset. Concentration units are ( $ng/m^3$ ) for all elements.

Element	Sources						Sum	Model/ Measured	
	Sea	Soil	LRT	Cu/Zn	Pb	Combustion	Model		
Al	0.00	7.10	0.00	1.08	1.85	3.14	13.17	38.70	34.0%
Si	4.45	27.31	0.27	10.80	7.75	2.76	53.35	69.50	76.8%
P	1.34	9.87	0.27	0.00	0.16	0.00	11.64	15.60	74.6%
S	0.41	0.00	32.49	40.42	0.00	165.08	238.40	396.60	60.1%
Cl	572.20	0.00	16.22	0.00	0.00	0.00	588.42	755.90	77.8%
K	14.91	9.14	25.42	0.00	9.94	15.07	74.48	76.00	98.0%
Ca	15.87	4.46	0.00	10.09	2.91	0.34	33.67	35.80	94.0%
Ti	0.40	0.37	0.00	1.12	0.35	1.10	3.33	3.40	98.1%
V	0.00	0.07	0.20	0.00	0.08	0.09	0.43	1.50	28.9%
Cr	0.33	1.06	0.14	0.07	0.00	0.22	1.82	3.70	49.2%
Mn	0.03	0.23	0.00	0.71	0.20	0.00	1.16	1.90	61.3%
Fe	1.26	8.66	0.60	20.69	5.23	1.69	38.12	37.90	100.6%
Ni	0.00	0.08	0.12	0.04	0.04	0.09	0.36	1.20	30.3%
Cu	0.00	0.01	0.00	0.95	0.24	0.22	1.41	2.40	58.9%
Zn	0.00	0.00	4.19	4.59	0.08	0.00	8.86	10.40	85.2%
As	0.03	0.00	0.17	0.28	0.04	0.24	0.76	0.80	94.7%
Se	0.03	0.01	0.06	0.00	0.00	0.02	0.12	0.10	116.9%
Br	0.84	0.23	0.65	0.23	0.00	0.23	2.17	2.10	103.5%
Rb	0.04	0.00	0.15	0.04	0.02	0.00	0.24	0.10	239.1%
Sr	0.30	0.01	0.06	0.00	0.02	0.00	0.38	0.60	63.2%
Pb	0.01	0.00	1.60	0.02	3.35	0.00	4.99	4.70	106.1%

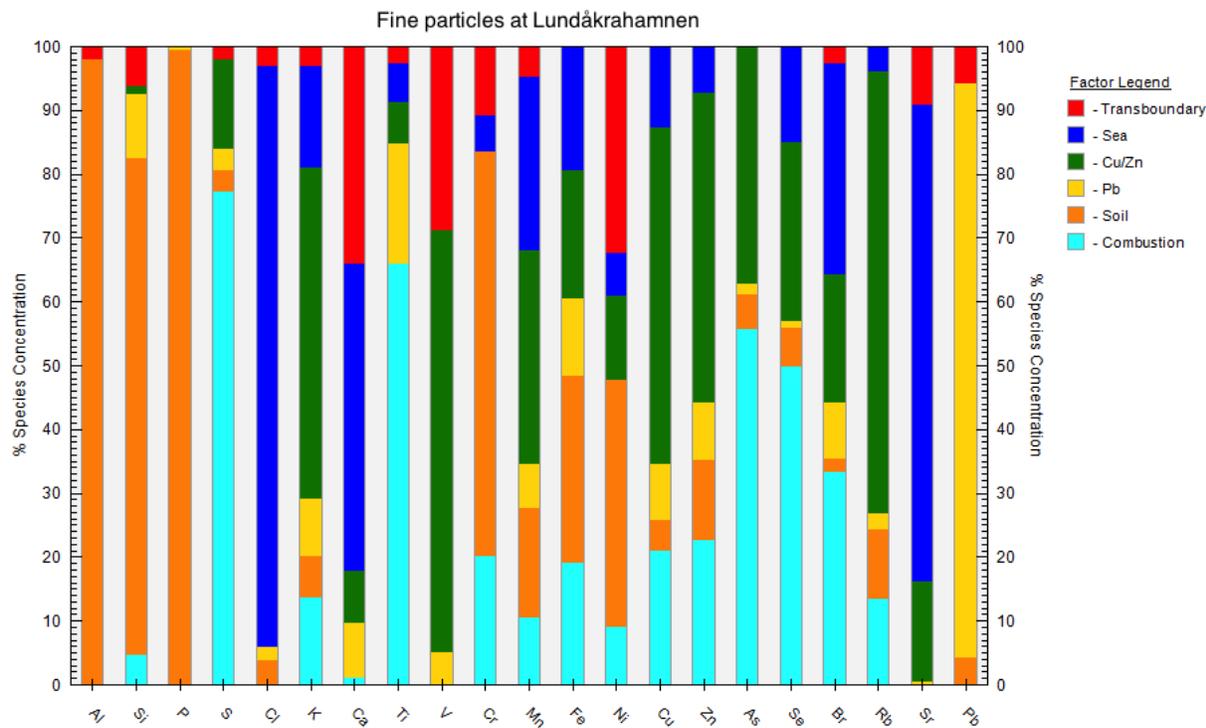
**Table 12:** Source apportionment according to the PMF model of fine particles at Lundåkrahamnen. Concentration units are ( $ng/m^3$ ) for all elements.

Element	Sources						Sum		Model/ Measured
	LRT	Sea	Cu/Zn	Pb	Soil	Combustion	Model	Measured	
Al	0.28	0.00	0.00	0.00	13.63	0.00	13.91	30.30	45.9%
Si	1.88	0.00	0.37	3.09	23.95	1.41	30.70	96.10	31.9%
P	0.00	0.00	0.00	0.04	8.75	0.00	8.79	21.10	41.7%
S	5.68	0.00	38.92	9.02	9.90	215.00	278.52	325.80	85.5%
Cl	11.39	350.44	0.00	8.58	14.52	0.00	384.93	410.60	93.7%
K	2.67	13.75	44.76	7.75	5.62	11.74	86.30	85.70	100.7%
Ca	10.55	15.01	2.49	2.71	0.00	0.35	31.10	31.40	99.0%
Ti	0.02	0.06	0.06	0.17	0.00	0.61	0.93	0.30	309.1%
V	0.28	0.00	0.64	0.05	0.00	0.00	0.97	1.00	97.1%
Cr	0.44	0.23	0.00	0.00	2.57	0.82	4.05	4.10	98.7%
Mn	0.10	0.60	0.73	0.15	0.38	0.23	2.20	2.30	95.8%
Fe	0.00	10.66	11.09	6.76	16.21	10.57	55.29	56.70	97.5%
Ni	0.30	0.06	0.12	0.00	0.36	0.08	0.92	1.60	57.5%
Cu	0.00	0.16	0.68	0.11	0.06	0.27	1.30	1.70	76.4%
Zn	0.01	0.65	4.43	0.81	1.16	2.07	9.12	10.10	90.3%
As	0.00	0.00	0.21	0.01	0.03	0.32	0.58	0.70	82.4%
Se	0.00	0.03	0.06	0.00	0.01	0.11	0.22	0.20	109.2%
Br	0.07	0.85	0.52	0.22	0.05	0.86	2.57	2.80	91.6%
Rb	0.00	0.01	0.13	0.00	0.02	0.03	0.19	0.20	94.0%
Sr	0.04	0.30	0.06	0.00	0.00	0.00	0.40	0.40	100.3%
Pb	0.99	0.00	0.00	15.58	0.74	0.00	17.32	17.50	98.9%



**Figure 22:** The factor fingerprint for fine particles at Stadshuset.

In Figure 23 the source profile for fine particles at Lundåkrahamnen has been presented. It can be seen that the two main metal sources are divided into Pb and Cu/Zn. By adding the sixth source to the PMF modelling parts of the Ni and Ca concentration were able to be explained by long range transport of pollutions. Elements that are emitted by the combustion source according to the PMF are S, Ti, As, Se and partly Br. S is typical for combustion but Ti should be related to Soil dust and Br is an element found in the sea. It is important to notice the uncertainty in the PMF results but to remember the overall conclusions. There is a source that emits Pb in amounts that are not comparable to the amounts measured from soil dust. The same conclusion can be made for the metal source that emits both Cu and Zn.



**Figure 23:** The factor fingerprint for fine particles at Lundåkrahamnen.

For the fine particles at both Stadshuset and Lundåkrahamnen there were 6 sources found. The sea source was characterised by both Cl and Sr while it also contained some soil elements. The soil source mainly contains AL, Si and Fe which are typical elements but this source also contains amounts of V and Ni. Those elements are typical for marine traffic but as it does not contain Cl from the sea, this is contradictory. At Lundåkrahamnen the soil source contains amounts of Pb. The two main sources of heavy metals contributing to fine particles are Cu/Zn source which mainly consist of Cu and Zn concentrations and the Pb source with mainly Pb concentrations. The combustion source contains mainly S, but as sulphur in fine particles mostly originates from condensation of sulphuric acid onto nanoparticles during long-range transport. This makes the combustion source more likely a transboundary source. Finally the LRT source contains high amounts of V and Ni which corresponds to marine traffic. This source also contains elements specific for the sea which makes it more a combustion/sea source.

To improve the source profiles achieved with the PMF model and to make the model/measured ratio closer to 100%, there are multiple options. First off, the model could be tested with different amount of sources. This could possibly model the contributions to certain elemental concentrations with a higher ratio. Another way of improving the source profile is by testing different FPEAK values. Higher polarisation could separate the sources more from each other, removing the contribution from the soil dust which exist in most sources. One could also try filtering out certain element concentration depending on their source to noise ratio. Testing different blanks could possibly affect the PMF model whereas certain elemental relationships are not maintained when using the lowest concentrations for some elements as was done in this project. One final option to improve the PMF model and

remove the soil dust contributions from all sources is by locking a source profile. This means that one lock a profile, so some elements are only allowed in that certain source.

To summarize this chapter, the found sources contributing to the airborne particles in Landskrona are:

**Source soil dust** The elements that identify this source are characteristic for soil dust (Al, Si, Fe).

**Source sea spray** The elements that identify this source are characteristic for sea spray (Cl, Br and Sr). High concentrations of Cl were measured due to winds from sea and higher wind speeds than previous campaigns.

**Pb-source** The Pb-source is mainly identified with element Pb. The wind directions recorded for the greatly increased Pb concentrations at Lundåkrahamnen and Stadshuset make Boliden Bergsöe AB most likely the source.

**Cu/Zn-source** The elements that identify this source are Cu and Zn. The local source that corresponded to these emissions and lies in the wind direction for the greatly increased concentration measured is the shipyard. For the lower concentration of Zn and Cu measured different sources are responsible, like boat paint and the motorway.

**Source combustion/sea** The elements that identify this source are characteristic for combustion of bad oil (V and Ni). Marine traffic is a possible contributor as the source profile also contains amounts of sea spray elements.

**Long-range transport** For the fine particles at both Lundåkrahamnen and Stadshuset, long-range transport was required to explain parts of the Pb and Ni concentrations. The elevated concentrations when local sources were not involved, were explained by emissions from the Copenhagen area by long-range transport. The same trajectories did pass Öresund and could be affected by marine traffic.

Compared to previous campaigns, no obvious source emitting Zn/Cr/Ni is found with the PMF model. In the previous campaign in 2008, Scandust AB was suggested as the Zn/Cr/Ni source. This lack of source emitting elements like Zn/Cr/Ni can be explained by the production stop of Scandust AB. However a new source emitting Cu and Zn is discovered. The same source also shows to emit As concentrations. The previous reports did mention a local source contributing with Cu concentrations and guessed it to be originating from the shipyard. This does seem like a correct conclusion together with the wind analysis.

## 6 Conclusion

The air quality in terms of heavy metal concentrations has been shown in both this study and previous studies to be poor in Landskrona. Elevated concentration levels of Pb, Zn, As and Cu were measured on several occasions at both Lundåkrahamnen and Stadshuset. In comparison to previous campaigns, no downward trend can be seen in the elemental average concentrations. On the contrary compared to the average concentrations from 2008, an increase can be seen but this is probably due to different wind direction, with more from the sea.

The average concentrations for the heavy metals are highly affected by a few samples with elevated values. The elevated concentrations do in some cases almost exceed the annual average limit set in the environmental quality standards but are in most cases far below. Although the average Pb concentrations measured are not alarming, a chronic exposure including several concentration peaks a year could affect human health. Chronic Pb exposure decreases children's learning ability and could lead to criminal behaviour at a later age.[30]

Depending on the wind directions, sources contributing to the aerosols in Landskrona are both local and transboundary. From the measurements and PMF modelling it shows that heavy metal concentration originate from mostly two sources. Boliden Bergsöe AB is the most probable source of Pb emissions at Lundåkrahamnen and a Cu/Zn source is traced to either Scandust AB or the shipyard.

To work towards a cleaner air in Landskrona, cooperation is required between both the industries and the environmental health department. The measurements should provide a base for more directed solutions. Regular measurements would favour further developments and stricter EQS could minimize emissions.

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# Appendices

## Appendix A - Statistical concentration data

In Table 13 to 16 different concentration values for the elements at Lundåkrahamnen and Stadshuset in the fine and coarse particles are presented.

**Table 13:** Different concentration values for elements in coarse particles at Lundåkrahamnen, ( $ng/m^3$ )

Element	Average	Median	Standard deviation	10th percentile	90th percentile	Max	Min
Al	32.6	23.6	38.4	5.8	70.7	167.1	0
Si	153.4	140.2	122.6	11.5	302.5	463.3	0.2
P	11.6	4.1	16.1	0	33.5	57.7	0
S	232.9	131.7	265.7	50.1	388.7	1289.5	19.7
Cl	1714.7	1024.2	1854.2	51.8	4983.8	6961.3	23.1
K	67.9	44	45	24	139.8	166.5	13.4
Ca	125.9	70.3	122.4	27.7	210.4	511.2	12.5
Ti	2.6	1.4	4.7	0	9.1	14.8	0
V	0.4	0.5	0.8	0	1.1	1.9	0
Cr	1	0.6	1.7	0	3.5	4.3	0
Mn	1.3	0.4	2.3	0	4.6	8.2	0
Fe	98.7	55	93.7	23.2	258.5	324.4	9.2
Ni	0.7	0.7	0.6	0	1.2	2.2	0
Cu	2.1	0.7	3.2	0.1	5.1	13.9	0
Zn	7.5	4.1	8.3	1.1	17	32.6	0
Ga	0	0	0.1	0	0	0.1	0
Ge	0	0	0.1	0	0.1	0.2	0
As	0.3	0	0.9	0	0.7	4	0
Se	0.1	0.1	0.1	0	0.3	0.4	0
Br	4	2.3	4.4	0.5	11.1	16.4	0
Rb	0	0	0.3	0	0.4	0.6	0
Sr	1.2	0.9	0.8	0.3	2.8	3.6	0.2
Y	0	0	0.2	0	0.2	0.3	0
Zr	0.2	0	0.7	0	1.3	2.3	0
Pd	1	0.8	0.9	0	2.6	3.1	0
Cd	0	0	2.2	0	0.8	3.8	0
Sn	3.8	2.7	4.4	0	9.2	15	0
Ba	0	0	19.9	0	0	25.7	0
Pb	24.4	0.6	89.3	0	6.6	418.1	0

**Table 14:** Different concentration values for elements in fine particles at Lundåkrahamnen, ( $ng/m^3$ )

Element	Average	Median	Standard deviation	10th percentile	90th percentile	Max	Min
Al	30.3	16.3	34.6	5.3	90.7	123.9	0
Si	96.1	59.9	112.4	20.4	153.2	591.5	14.8
P	21.1	9.3	22	1.5	53.4	68.2	0.2
S	325.8	240.4	264.7	45.1	739.6	928.7	4.3
Cl	410.6	150.1	665.3	4.3	1013.3	2854.8	0.7
K	85.7	73.7	49.8	27.3	154.5	200.9	6.2
Ca	31.4	21.2	35.1	4	68.6	166.2	0
Ti	0.3	0	2.8	0	2	12.1	0
V	1	0.7	1.1	0	2.4	4.4	0
Cr	4.1	3.1	3.9	0.3	10	12.8	0
Mn	2.3	2	1.4	0.7	4.3	5.4	0.2
Fe	56.7	58.3	32.6	17.9	91.8	126.6	0
Ni	1.6	1.5	0.9	0.4	2.8	3.7	0.1
Cu	1.7	1.2	2.3	0.3	3.1	12.1	0
Zn	10.1	7.8	8.8	2.4	24.5	37.9	0
Ga	0	0	0.1	0	0	0.1	0
Ge	0.1	0	0.2	0	0.3	0.6	0
As	0.7	0.3	0.9	0.1	1.6	4.1	0
Se	0.2	0.2	0.2	0	0.5	0.7	0
Br	2.8	2.5	2	1	5.5	8.1	0
Rb	0.2	0.1	0.3	0	0.6	0.8	0
Sr	0.4	0.2	0.6	0	0.8	2.4	0
Y	0	0	0.4	0	0.3	1.2	0
Zr	0	0	0.3	0	0.2	0.5	0
Pd	0	0	1.2	0	1.1	2.8	0
Cd	2.4	2	2.1	0	5.3	7.1	0
Sn	4	3.7	4.1	0	10.1	10.9	0
Ba	18.9	6.4	21.3	0	40.7	85.5	0
Pb	17.5	1.9	50	0.1	35.6	249.7	0

**Table 15:** Different concentration values for elements in coarse particles at Stadshuset, ( $ng/m^3$ )

Element	Average	Median	Standard deviation	10th percentile	90th percentile	Max	Min
Al	38.7	27.6	39.3	19.1	68	135.2	10.3
Si	69.5	65.6	136	17.6	138.8	160.1	7.5
P	15.6	4.3	14.3	0	45.7	49.7	0
S	396.6	242.8	80.1	114.8	716.8	2225.3	99.4
Cl	755.9	354.1	1451	88.8	1956.4	3134.1	34.6
K	76	58.6	25.6	34.2	128.4	325.3	17.9
Ca	35.8	32.9	125.2	15.6	71.8	85.6	3.4
Ti	3.4	2.3	6	1.2	6.8	16.5	0
V	1.5	1.4	0.9	0	3	4.4	0
Cr	3.7	2.8	2.4	0.8	6.8	12.6	0.2
Mn	1.9	1.4	1.9	0.6	4.4	5.9	0
Fe	37.9	18.1	127.9	8.1	84.5	153.3	0
Ni	1.2	1	0.5	0.7	1.9	2.7	0.5
Cu	2.4	0.7	55	0	4	28.9	0
Zn	10.4	4.7	25.3	1.8	27.9	54.3	0.5
Ga	0	0	0.1	0	0.2	0.3	0
Ge	0	0	0.1	0	0.2	0.3	0
As	0.8	0.4	0.3	0.1	1.5	6.3	0
Se	0.1	0.1	0.1	0	0.3	0.6	0
Br	2.1	1.8	3.4	0.3	4.3	7.1	0
Rb	0.1	0.1	0.3	0	0.6	1.2	0
Sr	0.6	0.4	0.8	0.1	1.1	2.2	0.1
Y	0.2	0.1	0.2	0	0.5	1	0
Zr	0	0	0.5	0	0	0.5	0
Pd	0.5	0.3	0.6	0	1.6	2	0
Cd	1.7	1.7	1.5	0	3.8	5	0
Sn	0.3	0	4.9	0	3.8	8.1	0
Ba	8.3	4	15.6	0	23.2	48.4	0
Pb	4.7	1	9.7	0	9.9	32.7	0

**Table 16:** Different concentration values for elements in fine particles at Stadshuset, ( $ng/m^3$ )

Element	Average	Median	Standard deviation	10th percentile	90th percentile	Max	Min
Al	67.4	66.7	29.2	27	113.2	159.1	8.1
Si	189	176.5	47.3	21.3	368.9	421.4	8.8
P	12.9	9.5	18.9	0	37.8	39.4	0
S	146.8	126.4	450.9	71.2	269.1	350.1	47
Cl	1787.5	1539	887.7	245	3887.8	5987.1	164.8
K	43.7	43.7	62.4	11.8	77.7	104.8	8.9
Ca	145.1	115.8	22.1	20.2	285.7	464.2	0
Ti	7.8	7.2	3.4	1.3	13.9	24.9	0
V	0.4	0.3	1.2	0	2	2.9	0
Cr	3.6	2.9	2.9	0.6	6.6	9.4	0.3
Mn	1.6	1.3	1.5	0	4.2	6.3	0
Fe	103.9	64	37.6	2.1	196.2	477.6	0
Ni	0.7	0.8	0.5	0.2	1.3	1.9	0
Cu	14.9	1.8	5.9	0.7	9.7	272.1	0.3
Zn	11.7	4.7	12.8	1.2	13.5	123.2	0.4
Ga	0	0	0.1	0	0	0.1	0
Ge	0.1	0	0.1	0	0.2	0.2	0
As	0.1	0	1.3	0	0.6	0.9	0
Se	0.1	0.1	0.2	0	0.2	0.3	0
Br	2.8	1.7	1.8	0.1	8.7	11.9	0
Rb	0	0	0.4	0	0.1	0.4	0
Sr	1	0.7	0.5	0.2	2.1	3.2	0
Y	0.2	0.2	0.3	0	0.4	0.6	0
Zr	0.4	0.2	0.3	0	1	1.5	0
Pd	0.4	0.1	0.7	0	1.5	1.8	0
Cd	1.5	1.1	1.6	0.1	2.9	6.2	0
Sn	4.4	3.8	3	0.2	7.5	24.4	0
Ba	11.8	7.8	13.3	0	34	59.6	0
Pb	4.9	1.4	8.7	0	18.3	41.3	0