Ship emissions contribution to particle concentrations in Falsterbo

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Abstract

The aim of this study is to estimate the average contribution of airborne particulate chemical substances in Falsterbo due to shipping emissions. Particularly the sulphate content will be analysed and its effect on both the environment and health will be discussed. The contribution was calculated using measurements in Falsterbo with a Soot Particle Aerosol Mass Spectrometer (SP-AMS) amongst other instruments. The SP-AMS measures particle concentrations of sulphate, nitrate, ammonium, organic aerosols, chlorine and refractory Black Carbon.

Based on data from one specific day, when the shipping contribution is high, the contribution from shipping to particulate sulphate was 0.88% of the total sulphate concentration, which corresponds to 0.007 $\mu g/m^3$ sulphate. For the entire measurement period, the sulphate contribution was between 0.4 and 0.5%.

The contribution of the other substances was overall low with only a few distinct peaks, even though ship plumes could be seen. This could be due to low emission factors of these compounds.

An analyse of data from the entire measurement period will be necessary to obtain a more accurate result.

The contribution of the author to this work

A service maintenance was done on a weekly basis during the entire period of measurements, including air flow checks and control of measurement data. If necessary, tube fittings were redone. After this, the majority of the time was spent on compiling the variety of data into one sheet in Excel with matching time resolution. Only a minor part of the time was spent on the data analysis. The remaining part of the time was dedicated for report writing.

Contents

1 Introduction

By causing changes in the Earth's atmosphere, in the amounts of greenhouse gases, aerosols and cloudiness, human activities contribute to climate change. Greenhouse gases and aerosols affect incoming solar radiation and outgoing thermal radiation, which in turn affect the climate. These changes can lead to a warming or cooling of the Earth's surface [1]. Among the anthropogenic species, aerosols cause the most adverse health effects through the inhalation in the human respiratory system [2].

1.1 Aerosols

An aerosol is a mixture of solid and liquid particles in a gas. The solid and liquid aerosol particles can vary in size, from ultrafine (< 100 nm diameter), to fine (< 1 μm diameter), and finally to coarse (1-10 μm diameter) [3]. The air that we breathe contains both natural and anthropogenic aerosol particles in these size ranges.

The anthropogenic atmospheric aerosol particles are affecting the climate through the direct and indirect effect. Aerosol particles are scattering or absorbing solar radiation. As human activity leads to an increase of aerosol particles in the air, the scattering of solar radiation back to space is increasing as well as the absorption of solar light. Thereby, this leads to a cooling of the earth surface. This is called the direct effect of the anthropogenic aerosol particles [3].

Particles also have an indirect way of affecting the climate through the alteration of cloud droplets. Namely, part of the atmospheric aerosol particles also act as seeds for cloud droplet formation, called cloud condensation nuclei (CCN). Due to anthropogenic emissions, the number of CCN increases in the atmosphere, which usually leads to optically thicker clouds and more sunlight being reflected back to space. Hence, this effect, called the first indirect effect is cooling. Also the lifetime of the cloud is increased, as the same amount of water can condense on more particles. Thereby, the droplets need more time to grow big enough to fall out as precipitation. This is called the second indirect effect [3].

The climate effect from the shipping sector may be relatively small compared to other modes of transportation, but the negative health effect is a major problem. Many studies have been performed showing a correlation between particulate matter and negative health effects when they are inhaled in the human respiratory system, such as asthma, heart attacks, and premature mortality [4]. Fine particles are assumed to be affecting human health the most of the three particle modes, since they include sulphates, nitrates, acids, metals and other chemicals that have been adsorbed onto their surfaces [2]. These particles can also for instance be breathed more deeply into the lungs and be transported over longer distances [5]. Ultrafine particles are believed to penetrate the vessels, via the alveolus, and cause or aggravate cardiovascular diseases [6].

1.2 Climate effects from shipping emissions

Combustion from ship engines is one source, which can affect climate. Ships mainly affect the climate through the emissions of nitrogen oxides (NO_X), sulphur dioxide (SO_2), carbon dioxide (CO_2), and particles containing black carbon (BC) [7].

The NO_X emissions result in a cooling effect by reducing methane lifetime over open ocean areas. NO_X also contributes to the formation of tropospheric ozone and shipping contributes 15-30% to global fossil fuel sourced NO_X emissions [4]. If there is excess ammonia and the sulphate aerosol is fully neutralised nitrate aerosols can form. The radiative forcing due to nitrate aerosols is negative [8].

Of global anthropogenic SO_2 emissions 5-8% originate from commercial shipping. The emissions of SO_2 lead to a cooling effect on the climate when sulphate particles are formed, either directly by scattering solar radiation, or indirectly by formation of CCN and cloud droplets [9]. Sulphate is formed through reactions within cloud droplets, or oxidation of SO2 and condensational growth onto pre-existing particles. Sulphate aerosols are almost entirely scattering and therefore contributing to a negative radiative forcing [10]. Lately, the shipping industry and organizations have attempted to reduce the impact on the environment through regulations. In January 2015, a new limit of the sulphur content in the fuel was set to 0.1 % for the Baltic Sea, North Sea, and the English Channel (SECA-area in Fig. 1). The decrease of the sulphur content in the fuel after the new regulations in 2015 can be seen in Fig. 2. A global limit of 0.5% sulphur content in fuel starting 1 January 2020 is expected [11]. Also NO_X emission limits in the SECA-area are being discussed for the year 2021.

Commercial shipping vessels represent approximately 3.3% of global anthropogenic emissions of CO_2 (excluding CO_2 from land use change) [12], which has the highest positive radiative forcing of all the human influenced climate drivers [13].

Black carbon absorbs solar radiation and heats the atmosphere. Above highly reflective surfaces, such as snow or clouds, it reduces the albedo of the surface and causes a positive radiative forcing [14]. Shipping contributes approximately 2% to global BC emissions [15].

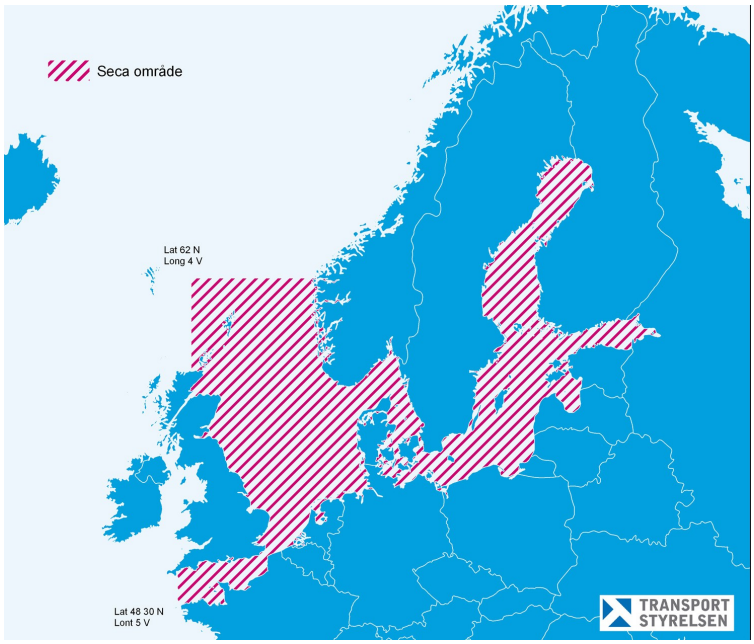

Figure 1. *The SECA-area marked in red [16]*

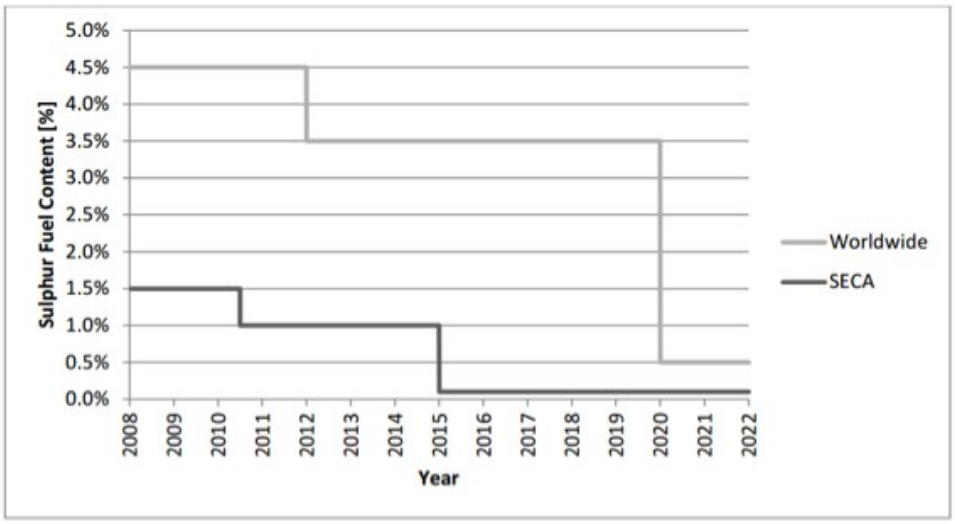

Figure 2. Sulphate limits for the Seca-area and worldwide

1.3 Radiative forcing from shipping emissions

Radiative forcing is the net irradiance at the tropopause and is used to compare the anthropogenic and natural drivers of climate change. Aerosols can directly cause a positive or negative forcing. Indirectly they cause a negative forcing [13]. The net radiative forcing due to ship emissions and the effects of ozone, reduced methane lifetime, sulphate particles, CO_2, and BC (Fig. 3a), is negative, and equal to approximately -70mW/m^2 (Fig. 3b). This forcing is relatively small compared to the solar constant of approximately 1.36kW/m^2.

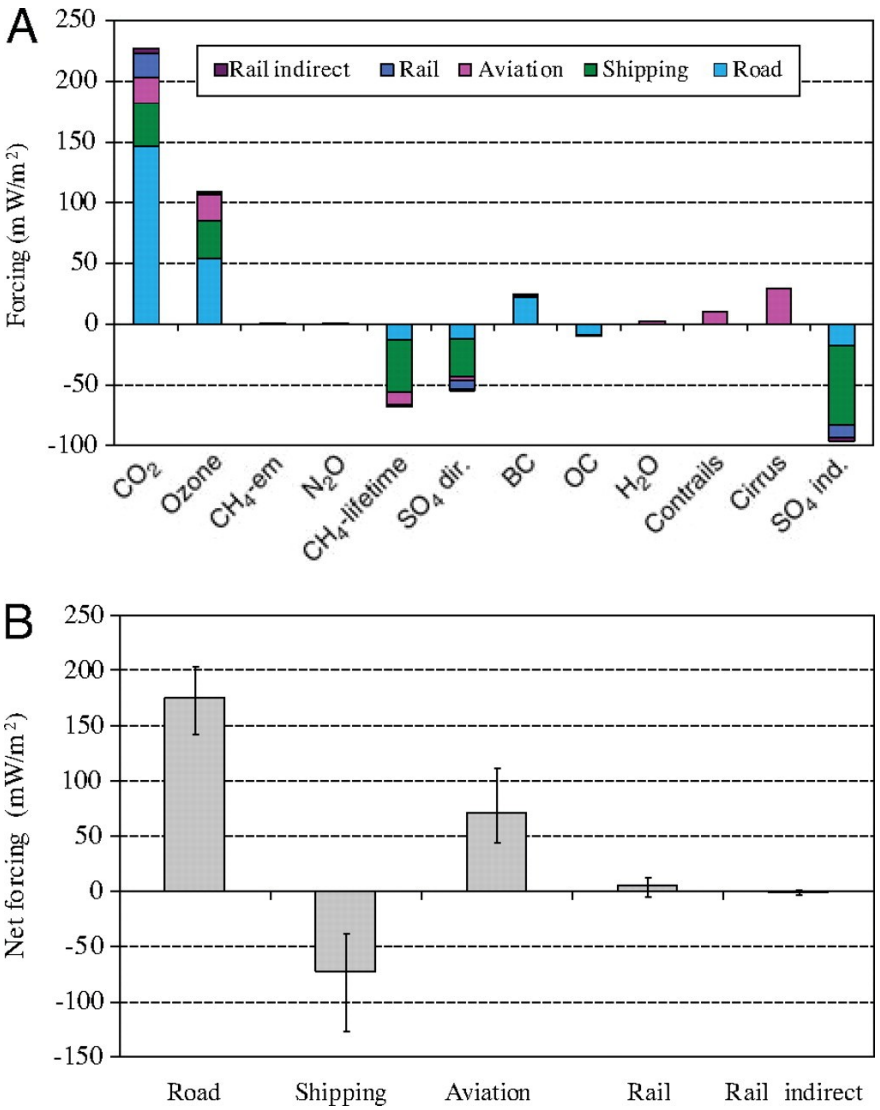

Figure 3. *(a) The radiative forcing due to different transport sectors and different gaseous and particle emissions, and (b), the net radiative forcing. [17]*

1.4 Health effects of shipping emissions

A study by Corbett et al. (2007), using ambient PM concentration models and population models, estimates that shipping emissions contribute to nearly 60,000 deaths globally each year, with the greatest impact at the coastal areas. The main reason for the large effect of shipping on health is that 70 % of shipping occurs within 400 km of land [18-19]. Most casualties have been estimated for Europe and Asia, where high populations and high shipping-related particulate matter concentrations coincide [20]. In Fig. 4 the annual cardiopulmonary mortality due to shipping emissions is estimated for Europe/Mediterranean, with most cases seen near the coastal regions.

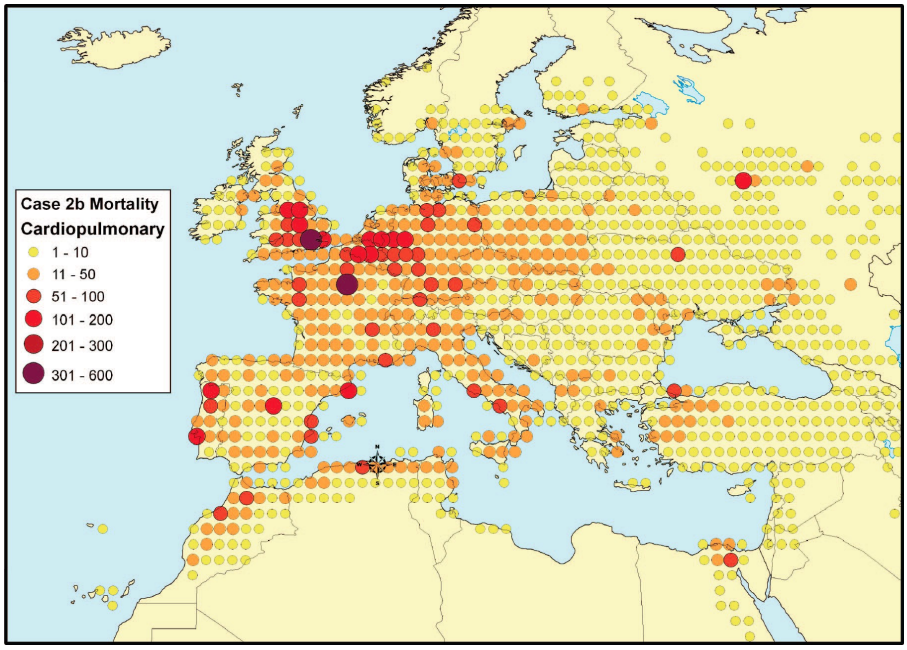

Figure 4. *The annual cardiopulmonary mortality due to shipping emissions for Europe/Mediterranean [4]*

2 Experimental

2.1 Measurement site

A station with measuring instruments was set up in Falsterbo to measure the ship emissions from mid January until the beginning of March. The measurement wagon (Fig. 5) and the location of the station (Fig. 6) can be seen below. The travel time for the emissions to reach the station is approximately 30 minutes with westerly winds (the closest shipping lane is influencing air pollution in the wind sector from south to north-west of the station). The wagon is positioned about 250 m from the sea shore to the west, and another 1 km from the sea shore to the south. The sea shore line around the wagon is almost flat, with an elevation of roughly 0.5 m above sea surface altitude. There are cars passing on a road located ten meters to the west of the wagon, between the station and the sea shore. Also, tractors were working north of the station. These activities are affecting the measurements, but data from these sources have been filtered out. There are only a few trees and a lighthouse to the south-west of the wagon, otherwise, the land is covered by low grass between the sea shore and the wagon to the west and north-west of the wagon. The eastern wind sector is dominated by a golf course up to a few hundred meters away.

***Figure 5**. The measurement wagon in Falsterbo*

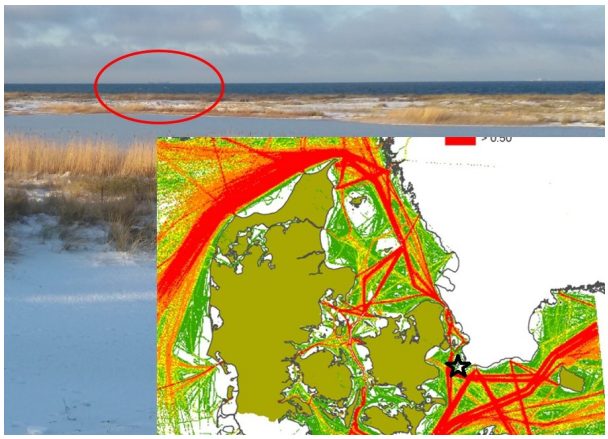

***Figure 6.** The location of the station in Falsterbo and a glimpse of a ship*

The total inflow of air to almost all the instruments was set to 16.7 litres/minute through a standard PM10 inlet and every instrument had its own fixed value of inflow, which can be seen in the flow scheme in Fig. 7. Only two instruments were disconnected from this flow, the SO_2 and NO_X monitor as seen in Fig. 7.

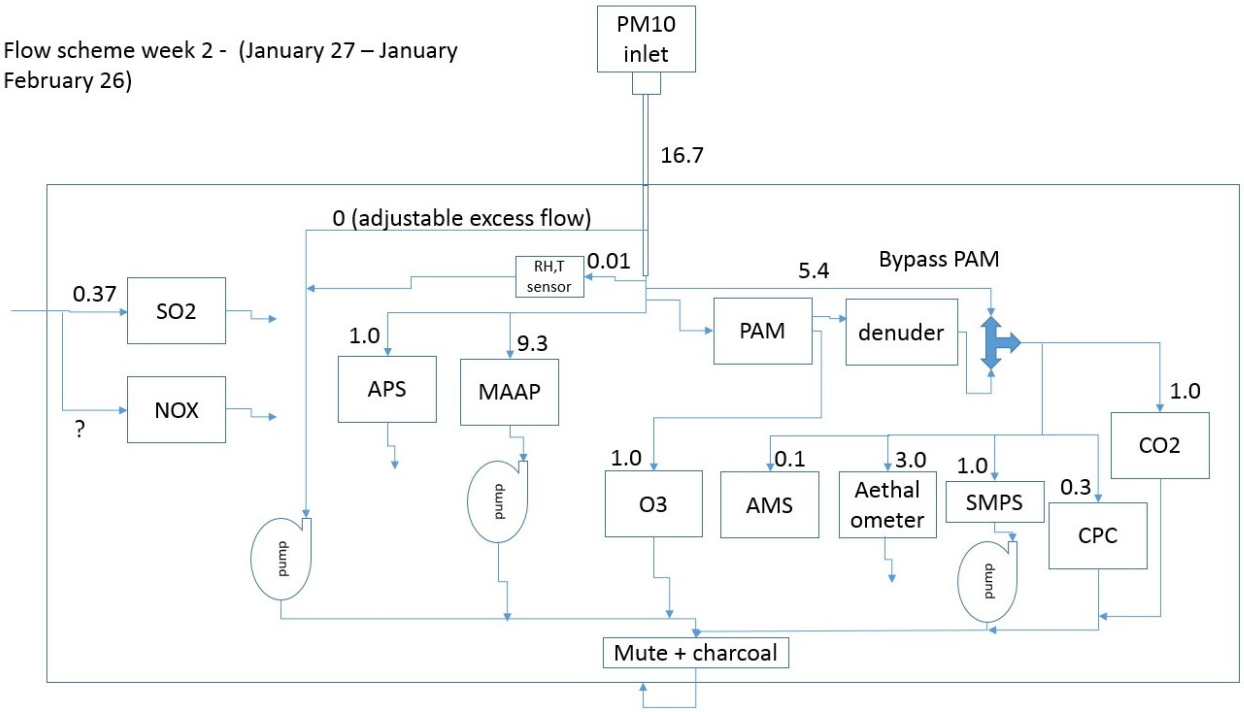

***Figure 7.** Flow scheme showing each instrument and its inflow of litres of air per minute (description of acronyms in section 2.2)*

2.2 Instruments

The instruments used at the station were; Soot Particle Aerosol Mass Spectrometer (SP-AMS), Aethalometer, Condensation particle counter (CPC), Scanning Mobility Particle Sizer (SMPS) spectrometer, Multiangle Absorption Photometer (MAAP), Aerodynamic Particle Sizer (APS) spectrometer, Potential Aerosol Mass (PAM) chamber, SO_2 monitor, NO_X monitor and a CO_2 monitor. The SP-AMS measures sulphate, nitrate, ammonium, organic aerosols, chlorine and refractory black carbon (rBC) (see sec. 2.2.1). The Aethalometer measures the mass concentration of BC particles passing the filter with the inflowing air. With the CPC, the number of particles are counted (see sec. 2.2.3). The SMPS and APS spectrometers measure the particles size distributions between 6 and 600 nm (see sec. 2.2.2) and 0.6 to 20 μm diameter respectively. MAAP measures BC and aerosol light absorption properties. The PAM simulates atmospheric oxidation processes. Each of the monitors for SO_2, NO_X and CO_2 measure the concentrations of these chemical compounds.

2.2.1 SP-AMS

The SP-AMS measures the chemical and physical properties of nitrate, sulphate, ammonium, chlorine, and organic containing particles and rBC. In the standard AMS configuration, aerosol particles are thermally vaporized at approximately 600ºC. The vapour is then ionized with an electron beam and the ions are detected in a high-resolution mass spectrometer [21]. In this mode, the AMS is not detecting rBC [22]. For the rBC fraction, an intracavity laser vaporizer is added, which provides detection of both refractory and non-refractory components. If the particle material is characterised as refractory or non-refractory is dependent on the detection capability of the instrument [21,23]. Schematic views of the SP-AMS can be seen in Fig. 8. To optimize the laser performance, the laser cavity is located entirely within the AMS vacuum chamber. The SP-AMS has three orthogonal axes, consisting of particle beam, laser vaporizer and ion extraction.

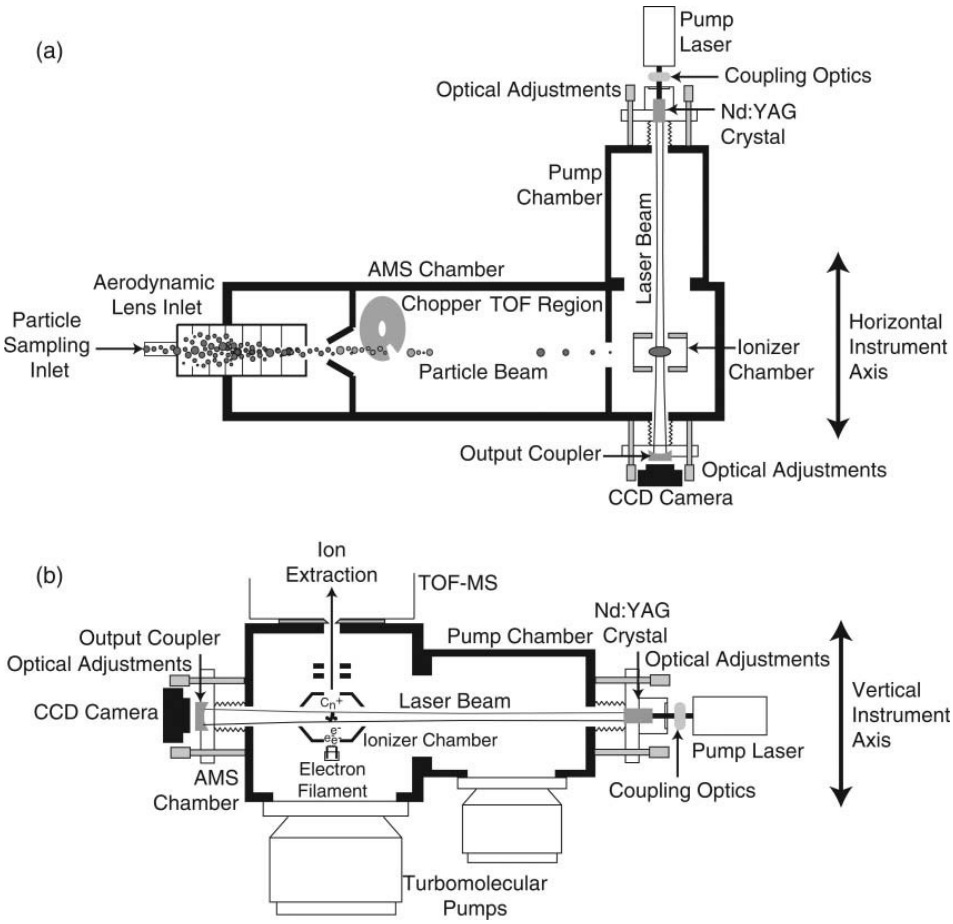

Figure 8. *A schematic view of the SP-AMS [14]*

Particles sampled into the vacuum chamber only spend a short amount of time in the laser beam due to their high speed and therefore high intracavity laser power is required. The particles scatter the laser light as they enter the laser beam, which can be seen as the red line in Fig. 9. Non-refractory material is vaporized from the particles, decreasing the scattered light, as the absorbing material in the particles starts to heat up. The rBC material continues to heat up further and the blackbody radiation at these temperatures gives rise to a visible incandescence signal (blue line in Fig 9). The vaporized species are ionized and detected in the SP-AMS [22].

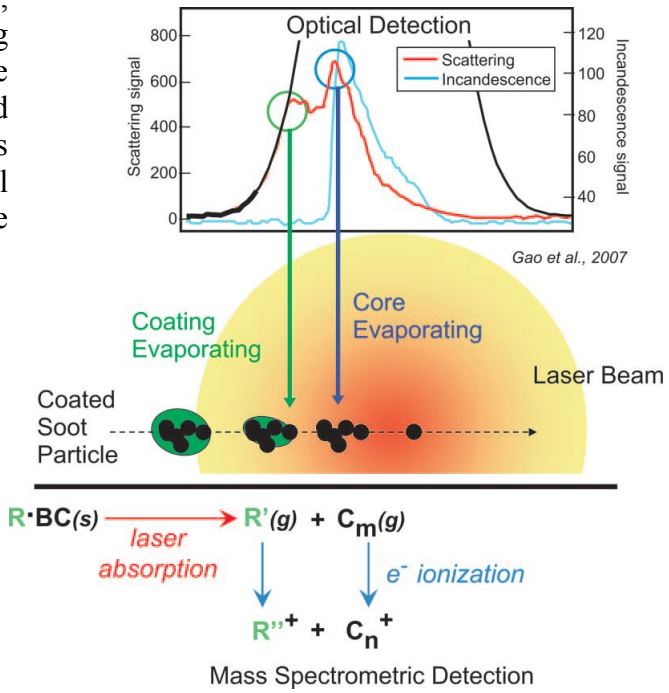

Figure 9. *Particle detection with SP-AMS [14]*

2.2.2 SMPS

The SMPS consists of a bipolar diffusion charger, a differential mobility analyser (DMA) and a condensation particle counter (TSI CPC 3010) [24]. The particles are first led through the charger, where they attain charges according to a well-defined charge distribution. When passing through the DMA the particles will be deflected due to the voltage applied. If the particles have the right electrical mobility they will be collected at the aerosol outlet slit (Qa in Fig. 10). If not, they will be transported away by the excess air flow or deposited on the walls. By applying different voltages the aerosol mobility distribution can be measured. By using an inversion algorithm and the CPC the aerosol number size distribution can be determined [25].

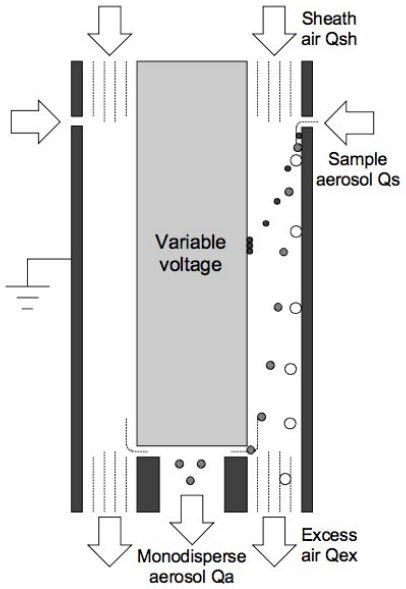

Figure 10. *A schematic view of the DMA [25]*

2.2.3 CPC

The CPC used for these measurements is a TSI CPC 3775. It detects particles down to 4 nm over a concentration range of 0-10^7 particles/cm^3. The incoming aerosol is mixed by the saturator (see Fig. 11) with Butan-1-ol saturated air. In the condenser the mixture is cooled so that the butanol vapour pressure is above the saturation vapour pressure. The particles then grow to a detectable size and are counted in the detection chamber by a laser diode and photo detector [26].

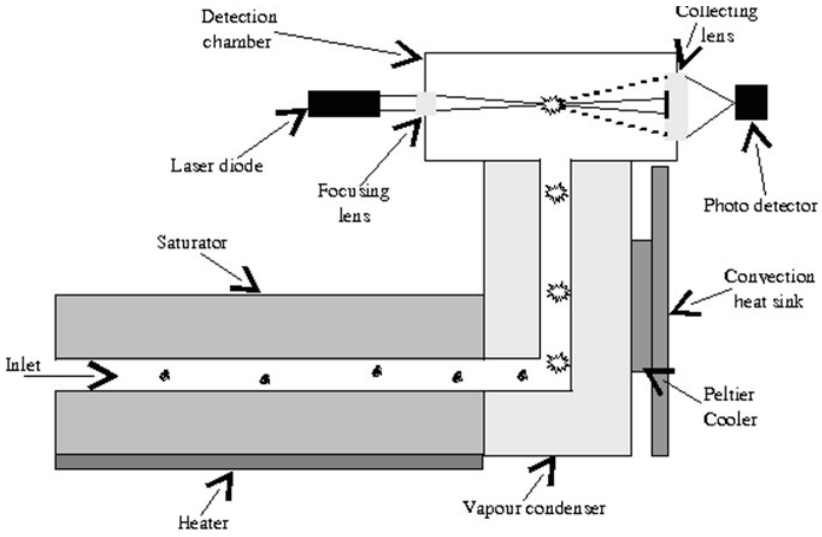

Figure 11. *A schematic picture of a CPC (copyright TSI. Inc.)*

3 Analysis method

All data used in this study is preliminary data. A calibration of the AMS was done before the measurements started, but the one performed during the measurement period has not been taken into account. The SMPS and CPC data has not been quality controlled.

In order to see the maximum contribution from the passing ships, days with northwesterly to southerly winds were selected by using SMHI's wind speed data from Falsterbo lighthouse. One of these days, namely, January 27 was chosen for the subsequent analysis. Data from AMS and CPC were plotted together and compared with SMPS plots, produced in Matlab, to identify ship plumes. Plumes that seemed to originate from other sources than ships, for instance from tractors working outside of the station, were ignored and not accounted for.

The number of ships passing Falsterbo during one day was estimated by counting the number of ships from a live map on the marine traffic website [27] during one hour and approximating the hourly number to be the same for the rest of the day. The amount of plumes were estimated from the plots and compared with the approximated “real” value.

To calculate the sulphate concentration from the ship emissions the peaks were divided into two categories: one with the peaks that are certain to be ship related and one with uncertain peaks. The time period studied is approximately 10.5 hours. A mean value of the background, measured by the SP-AMS, was calculated with the values of the concentration right before and after a peak. The background value was then subtracted from the sulphate peak. This was done for every peak and a daily mean value of the sulphate contribution, during days with right wind conditions, was estimated. Also an estimation of the percentage increase in sulphate concentration during the measurement period was done.

4 Results

4.1 Sulphate

A day with westerly winds was chosen to be analysed. The 27th of January was chosen and Fig. 11 is showing a plot for the first half of the day with data from the SP-AMS and CPC. Figure 12 is showing a plot from the SMPS for the same time period.

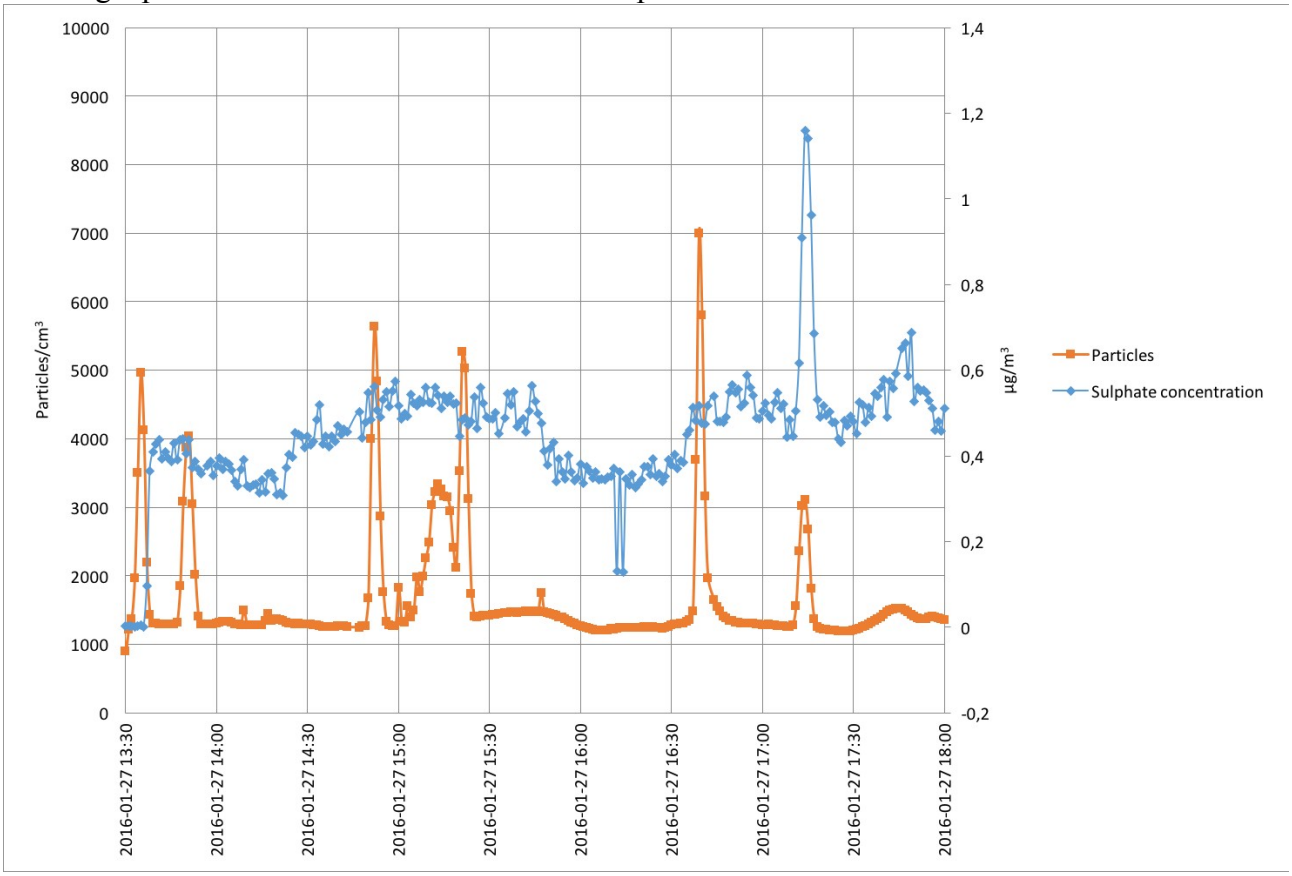

Figure 12. *Plot with data from the CPC and SP-AMS, showing total concentration of particles (/cm³) and the sulphate concentration (µg/m³) between 13:30 and 18:00 of the 27th of January*

Fig. 12 was compared with Fig. 13 to identify ship plumes. One clear sulphate plume has been identified in Fig. 12 just before 17:16. Every second hour, the soot module was turned on in the SP-AMS. When the module is on the measurements become more sensitive and therefore the background sulphate concentrations vary from higher to lower values every other hour.

The number of plume peaks with an elevated total particle number concentration is much higher than for sulphate. In Figure 13 the ship plumes based on the number concentration are shown as the more intense red coloured areas in the upper panel at particle diameters between roughly 10 and 100 nm diameter. There is also a ship plume, which has the highest number concentration registered for sizes between 50 and 200 nm diameter around 17:20.

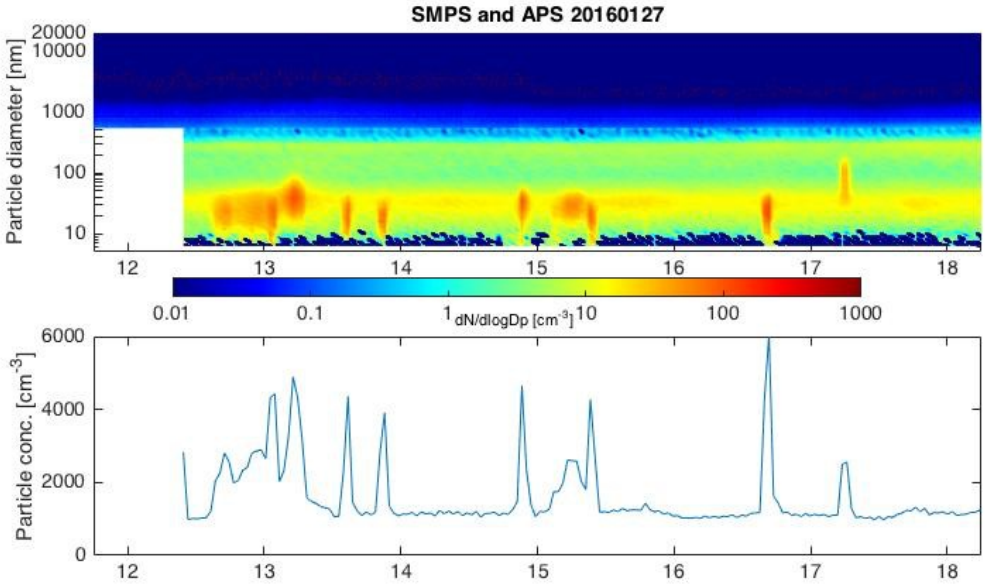

Figure 13. *Plot from the SMPS particle number size distribution (upper panel) and total particle number concentration (/cm^3) (lower panel) during January 27, 12:30-18:20*

Fig. 14-15 are showing the measurements for the last part of the 27th of January. With two certain sulphate peaks originating from ship emissions.

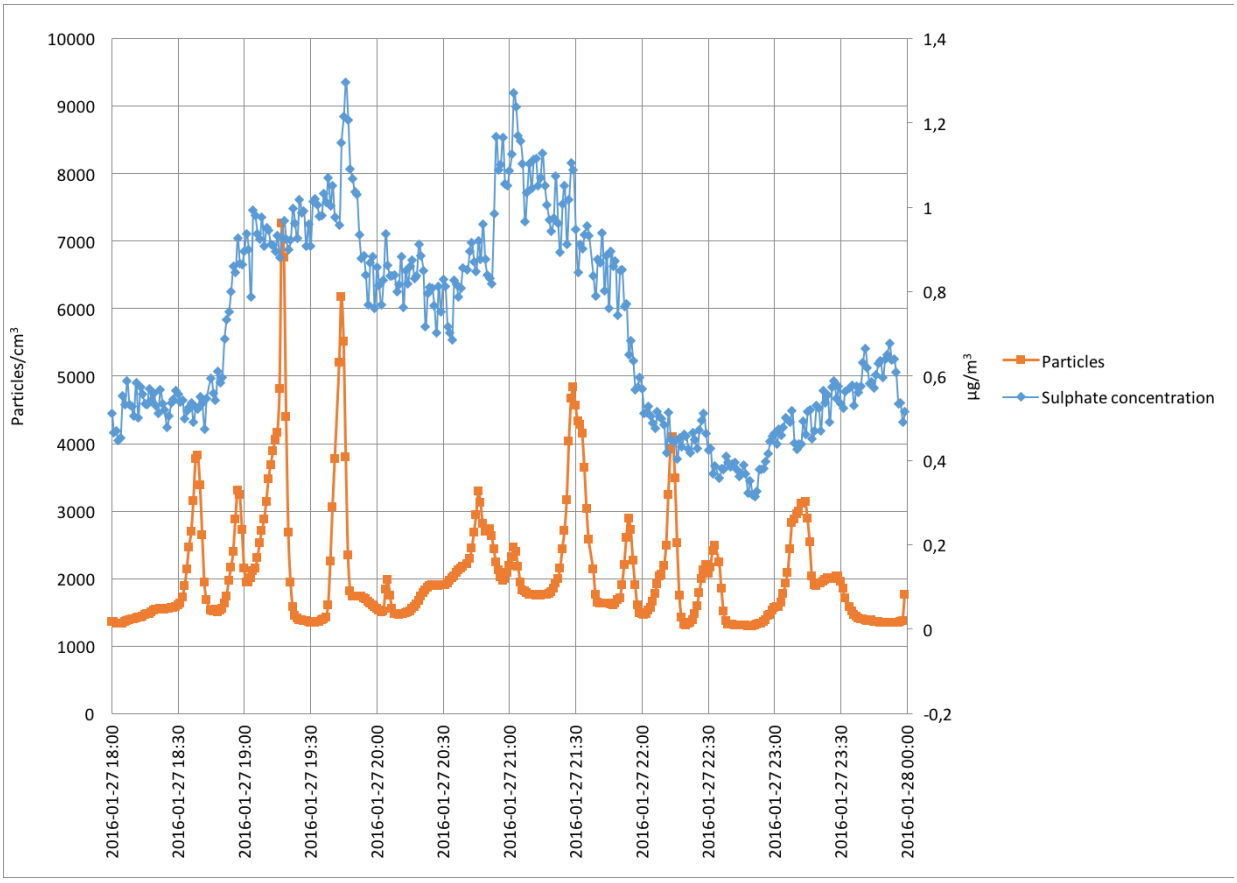

Figure 14. *Plot with data from the CPC and SP-AMS, showing total concentration of particles (/cm³) and the sulphate concentration (µg/m³) between 18:00 and 24:00 of the 27th of January*

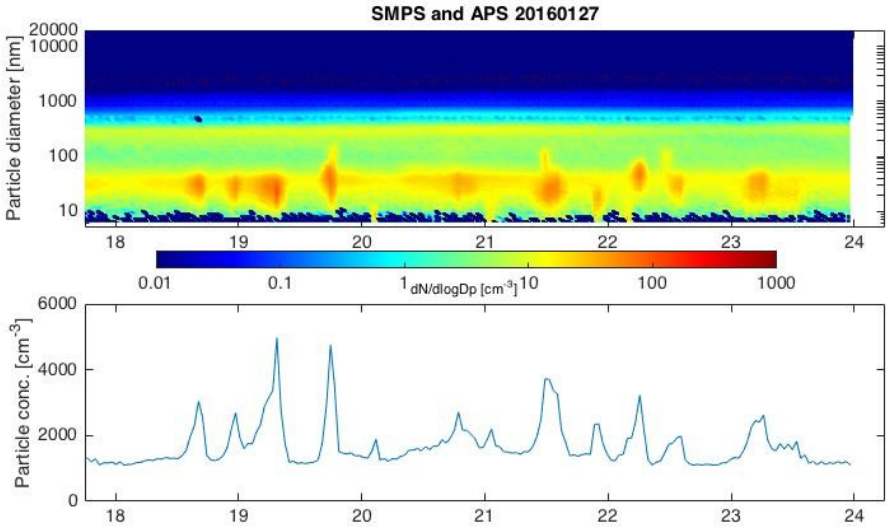

Figure 15. *Plot from the SMPS particle number size distribution (upper panel) and total particle number concentration (/cm³) (lower panel) during January 27, 17:50-24:00*

When the ship identification is based on the number concentration, eleven certain single ship plumes can be seen during January 27 and eight uncertain peaks that seem to contain two ship plumes each. This gives an estimated daily value between 25 to 62 ships passing per day which are contributing to elevated particle number concentrations. By looking at the marine traffic website five ships were passing during one hour, which gives an estimated daily value of 120 ships. Hence, at least half of the ships are polluting the environment less in the sense that these ships do not enhance particle concentrations above background levels.

Three certain sulphate peaks from ships are seen during this period and one that is uncertain. The average contribution from each of the certain peaks is 0.25 $\mu g/m^3$, which gives an average contribution of 0.007 $\mu g/m^3$ for the entire day. This corresponds to a contribution of 0.88% for this day. With the uncertain peaks included in the calculations the average contribution is 0.008 $\mu g/m^3$ for the entire day and a percentage contribution of 1.0%.

The total percentage of added sulphate during the entire measurement period was estimated. January 27 represents a typical day with winds from the west, when sulphate contribution from ships estimated to 0.88% is highest. There are other days with westerly winds, when the ship contribution is much lower. The contribution during days when the ship contribution reaches its minimum, has not been calculated, but should be around 0.4%. Hence, the ship contribution during days with westerly winds is somewhere between 0.4% and 0.88%. For days with north-westerly, or southerly winds, the contribution should be on the order of 0.3% to 0.8%. All in all, the average contribution to sulphate from ships when the winds are from south, west, or north-west, should be somewhere in between these two ranges, namely 0.35% to 0.84%, with an average contribution of 0.6%.

From SMHI's measurements at Falsterbo lighthouse, there are 64% instances with winds from south to west and northwest between January and March. With the assumption of a zero percentage sulphate contribution from ships during days with other wind directions, the estimated average percentage increase of sulphate during the measurement period is 0.4%. This is calculated with the following formula:

Average contribution = (64% x 0.60% + 36% x 0%) / 100% ≈ 0.4%

Hence, ships contribute to an increase of the sulphate concentration at Falsterbo by roughly 0.4% during the entire winter.

With the uncertain sulphate peaks taken into account the maximum percentage increase of the concentration is approximately 0.7% and would result in an estimated average percentage increase of 0.5% for the entire measurement period.

4.2 Other substances

Apart from sulphate, also organic aerosols, nitrate, ammonium and chlorine were being measured with the SP-AMS. Fig. 16 is showing a part of these measurements from the 27th of January. The ship contribution of the other substances is very low and is difficult to distinguish from the background, except for one small peak of organic aerosols that can be seen in the figure around time 14:44. When the soot module is on, every other hour, rBC is also being measured. Concentrations of rBC are shown as the black line in the figure, with no distinct peaks.

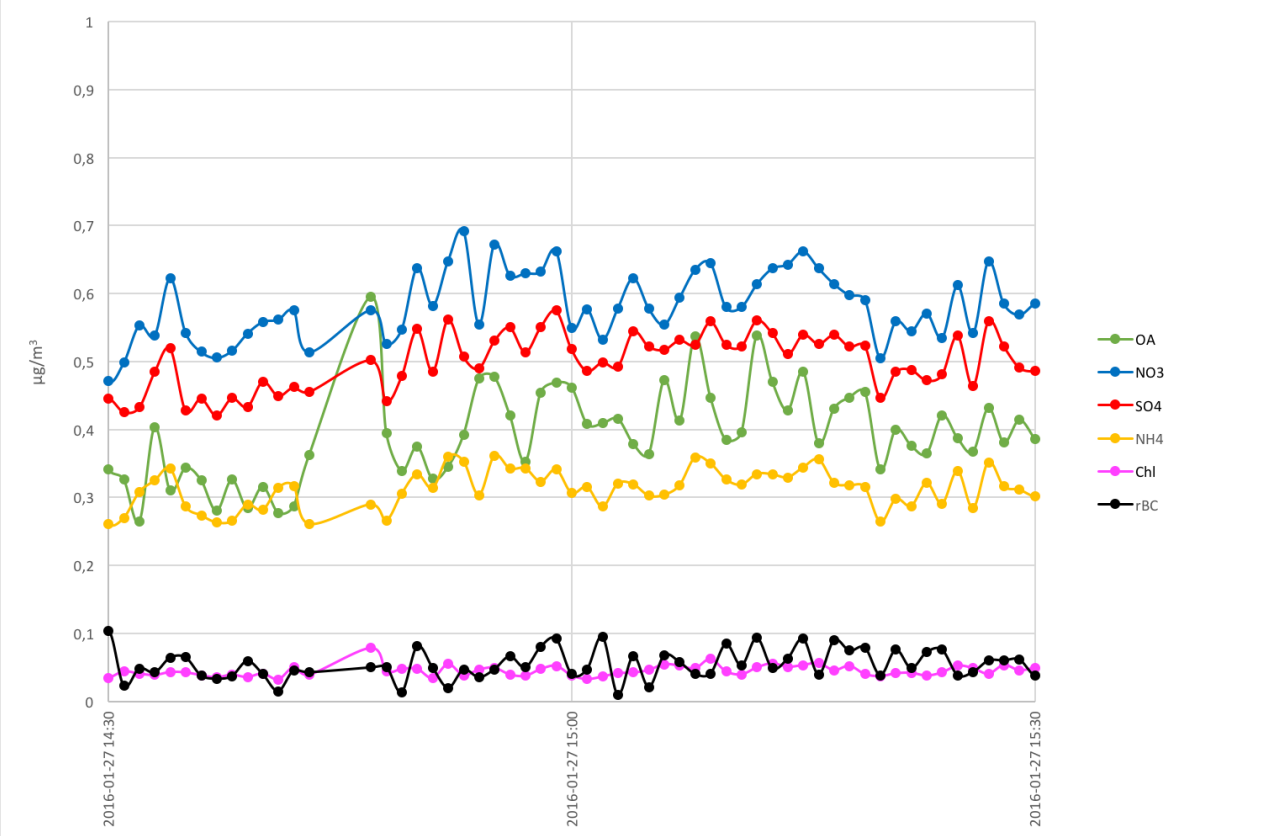

Figure 16. *Plot with data from the SP-AMS, showing concentrations (µg/m³) of organic aerosols, nitrate, sulphate, ammonium, chlorine and refractory black carbon between 14:30 and 15:30 of the 27th of January*

5 Discussion and conclusion

The result shows an overall low contribution from ships to the substance concentrations. Also, very few ships contribute to high concentrations of sulphate. These results may only be representative for the winter season, due to different meteorological conditions and solar light intensity during summertime. The contribution to sulphate and other substances might be higher in summer as suggested by model results by Matthias et al. [28].

At other non-analyzed days there seems to be a peak in the sulphate concentration but only a very small peak from the CPC. This can be due to high amounts of sulphuric acid, emitted from the ships, condensing onto large particles and thereby not allowing for the production of new nanometer sized particles. If the large particles are exceeding 100 nm in diameter, this condensation could be common. The greater the concentration of particles at larger diameters, the higher the particle surface available for condensation.

Since almost 50% of PM measured within a few minutes of the emissions is a result of fuel sulphur, the sulphur regulations discussed previously are expected to have a major impact on the PM concentrations. For every 1% increase in fuel sulphur there is a 13% increase of PM that can act as CCN. Even though PM concentrations are reduced with lower sulphur content, PM lifetime might increase since less particles will form cloud droplets. Longer lifetime of an aerosol will have a larger impact on the radiative forcing [11].

Also organic aerosols are dependent on the sulphur content in the fuel. With lower sulphur content their concentrations are expected to decrease [11], which corresponds well to the plots analysed. Not many peaks of organic aerosols could be seen in Falsterbo, where fuel sulphur content is regulated to 0.1%.

The emissions and particle composition are not only dependent on the sulphur content of the fuel, they are also affected by the engine type, vessel activity and maintenance [11].

6 Outlook

In order to improve the estimations of how much ships contribute to sulphate levels in this study, data from the entire measurement period would need to be analysed.

Also, a number of approximations have been made in the results. The minimum percentage sulphate increase and the percentage increase due to certain wind directions have only been estimated by observing the concentration graphs and by making rough calculations. More meticulous calculations of the background would be needed in order to get a more accurate result. The assumption that the contribution is zero from all other wind directions might not be valid.

The winds were assumed to be evenly distributed between southerly, westerly, and north-westerly wind directions. A more precise calculation of the wind directions could yield a slightly different value of the ship contributions.

The varying background concentration for varying wind directions has not been taken into account. For example, winds passing over Copenhagen before arriving at Falsterbo might significantly increase the contribution to background concentrations compared to an air mass that passed Denmark, but not Copenhagen.

In other words, given more time, a more precise calculation of the ship contribution to particle concentration could be made.

Acknowledgements

I would like to thank Adam Kristensson who let me join his project in Falsterbo and for sharing his knowledge of the subject.

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