

Interactive comment on "Methods for identifying aged ship plumes and estimating contribution to aerosol exposure downwind of shipping lanes" by Stina Ausmeel et al.

Stina Ausmeel et al.

stina.ausmeel@nuclear.lu.se

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We appreciate the comments by the referee, especially about wind uncertainties for our methodology, e.g. the impact of sea breeze, which was not explored in the manuscript. Now we have addressed this issue as described below.

Referee comment 1.

You can refer to two more studies in the literature that specifically focused on ship plumes and characterization of emissions in northern latitudes. These are given below and may be added to the literature review section: Aliabadi, A. A., Staebler, R. M.

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& Sharma, S. (2015), 'Air Quality Monitoring in Communities of the Canadian Arctic During the High Shipping Season with a Focus on Atmospheric Chemistry and Physics, 15(5), 2651-2673, doi: 10.5194/acp-15-2651-2015. Aliabadi, A. A., Thomas, J. L., Herber, A. B., Staebler, R. M., Leaitch, R. W., Schulz, H., Law, K. S., Marelle, L., Burkart, J., Willis, M. D., Bozem, H., Hoor, P. M., Kollner, F., Schneider, J., Levasseur, M., & Abbatt, J. P. D. (2016), 'Ship Emissions Measurement in the Arctic from Plume Intercepts of the Canadian Coast Guard Icebreaker Amundsen from the Polar 6 Aircraft Platform', Atmospheric Chemistry and Physics, 16(12), 7899-7916, doi: 10.5194/acp-16-7899-2016.

Author's response 1.

We acknowledge that we have not covered the complete literature when it comes to ship emission studies, since this is a large and wide subject (see also response to Referee 1, comment 1). The suggested papers are interesting. The first can be a good comparison between our results and a clean environment as the Arctic. The second paper presents airborne plume measurements, which we have not discussed in detail, but as is also mentioned in Referee Comment 4, the ship plume particle size distribution as a function of age is possibly comparable to some of the plumes in Falsterbo. All plumes in our manuscript are very fresh compared to the results in Aliabadi et al. 2016. We consider that the first paper suggested by the referee (Aliabadi et al., 2015) is the most relevant to our study and have chosen to include it in the section "1. Introduction section" together with several other papers relevant to our own work. There was a similar comment from Referee 1, and we have added a section in the manuscript which addresses the concerns of both referees.

Author's changes in manuscript 1.

"Particle number size distributions have been studied in atmospheric conditions previously, showing some variations in sizes and number of modes. This can be expected since many factors affect the emissions, such as engine operations, and the atmospheric transformation processes. For example, Jonsson et al. (2011) showed that size resolved particle number emission factors were largest around particle diameters of 35 nm, with smaller sizes observed for ships running on gas turbines than on diesel engines. Out of these particles, 36-46 % were non-volatile, and could contain some black carbon (BC). These measurements are from 2010, i.e. during the 1 % Sulphur limit within SECAs. Pirjola et al. (2014) showed that the number size distribution had two modes for fresh ship plumes, a dominating mode peaked at 20- 30 nm, and an accumulation mode at 80-100 nm. About 30 % of these were non-volatile, and it was also shown that the after treatment system affected the total particle number emission. These measurements are from 2010-2011. Diesch et al. (2013) observed a nucleation mode in the 10-20 nm diameter range and a combustion aerosol mode centred at about 35 nm. No particles with sizes above 1 μ m were observed. Six percent of the particle mass was due to BC. Other measurements on-board on a ship showed particle size distributions major peak at around 10 nm and a smaller peak at around 30-40 nm. Ca 40 % of the mass was non-volatile material, but particles below 10 nm consisted of only volatile material. (Hallquist et al., 2013) Westerlund et al. (2015) measured ship plumes from a stationary site and used AIS to characterise ships. Westerlund et al. found unimodal particle number size distributions for cargo and passenger ships, with the peak around 40 nm, while e.g. tug-boats emitted smaller particles. Since the measurements were carried out in a harbour area, as most of the other studies above, they could capture changes in emissions during e.g. acceleration of ships. These harbour measurements were carried out in 2010, i.e. also before the 2015 SECA implementation. In another harbour area, Donateo et al. (2014) quantified the contribution of ship emissions to local total aerosol concentrations. The ship contribution to particle number was found to be 26 %. They could also see plume peaks in PM2.5, since measurements were done in a harbour area and plume peak concentrations were relatively high. A study performed in an Arctic region, showed a size distribution mode with peak around 27 nm during the first 6 hours of plume transport and later (>6 h) modes above 100 nm become more prominent. (Aliabadi et al., 2015) Here, the ship contribution to

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BC was estimated to be 4.3-9.8 %. Due to the clean Arctic environment and low background concentrations, the evolution of a ship plume contribution could be studied over time (0-72 h). Dispersion modelling of ship plumes has shown that dilution and coagulation are important processes within the first hour after emission, reducing the number concentration by four orders of magnitude and one order of magnitude, respectively. (Tian et al., 2014) The decrease in particle number concentration is most rapid during the first minutes after emission."

Referee comment 2.

The most major concern is lack of accurate wind measurements. It is likely the air circulation patterns near coastal areas be very non-uniform horizontally. For instance wind speeds and directions can change significantly from the location of the ship to that of the weather station on land. I understand that the simplistic nature of the method justifies using a few weather stations, but the authors can investigate potential errors more. Below are some ideas. Was there wind speed and direction measurement on board of some ships? In this way you can characterize some differences between wind conditions on the sea and on land. You can also perform some hypothetical plume dispersion simulations near the coastal waters of interest to see if wind conditions are generally horizontally homogenous. You can use HYSPLIT web-based trajectory or dispersion modelling to investigate this quickly. For instance try some diurnal times and different seasons to investigate this. If you use trajectory modelling, you can investigate trajectories of air parcels arriving at the weather station of AQ trailer. Otherwise, if you use dispersion modelling, you can use point source and the ship stack to see where the plume goes. Having a few simple simulations included in the paper can add value on adequacy of the simplistic approach for meteorological model. (https://www.ready.noaa.gov/HYSPLIT.php)

Author's response 2.

The referee has raised a very important point about meteorology and winds, which af-

fects the applicability of the method, and we appreciate the two alternative suggestions to check how meteorology can affect our measurements. We have decided to follow the suggestions with air mass back trajectories. We argue that sea breeze situations are among the most extreme situations when wind measurements in an erroneous way can show that we have winds from a shipping lane, while in reality the air does not come from the shipping lane. During a sea breeze, the local wind direction can be reversed as compared to the large scale air flow. We decided to investigate sea breezes at our measurement site in Falsterbo, and also insert some recommendations in the manuscript for coastal measurements when sea breeze is a common phenomenon. During the development of a sea breeze, the winds close to the ground level at the shore line can slowly start to blow towards land due to land warming up more than the sea from solar light absorption, despite that the large scale circulation is showing a different wind direction. At the start-up phase, the winds closest to the shore line where we perform our wind measurements at our measurement station will then not agree with the winds over the shipping lane further away from the shore line. Hence, the ship plumes might not reach the measurement station despite that the measured wind direction is suggesting that. In this situation, our wind path method will not work, and we will not register an enhanced ship plume concentration at the station. The danger in this situation is that this is interpreted as if the ships are contributing negligible pollution to our measurement site, while in reality the winds from the ships have not even reached the station. On the other hand, with a fully developed sea breeze later in the afternoon, the horizontal extension of the winds going from the sea towards inland locations have increased and the ship plumes can potentially reach the measurement station again, and the method works once more. Fully developed sea breezes can have horizontal extensions on the order of more than 50 kilometers (Pokhrel and Lee, Atmospheric Pollution Research, 2011, 2, 106-115). Land breezes can also potentially be a problem for the analysis, although land breezes are normally weaker than sea breezes. When we checked if there were sea breeze situations in Falsterbo, we have discovered that indeed there are two different occasions (in total 4 days) when there

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were sea breezes developing during lunch time until late afternoon in May 2016. The local wind measured at the station indicated that we should be receiving air from the shipping lane to the west, while synoptic surface pressure wheather maps and our Hysplit trajectory analysis showed wind directions coming from the north. Unfortunately, we could not investigate how this affected the ship plume analysis at Falsterbo, since our instrumentation did not work during these sea breeze periods. There were no other strong sea breeze periods during the remaining period of our summer measurements. The sea breeze situations in Falsterbo could have disgualified our wind method, at least before the sea breeze became fully developed later during the day, and when the sea breeze started to fade out. With a continental area with a larger contrast in temperatures between land and sea and with larger continental and sea area, these problems can be even more common than in Falsterbo. There can also be other meteorological situations when the wind appears to be coming from the shipping lane, while in reality it is not. In summary, before performing ship plume measurements, each measurement location should be investigated for the occurrence of sea breezes and their horizontal extension, to be able to set up the experiments in a suitable way. However, the sea breeze problem and other meteorological phenomenon should not disgualify any measurement location. Namely, sea breezes do not take place all the time, and even during sea breezes, we might record shipping plumes at the shore line as we explained above. Nevertheless, users of the current method should be cautious to the occurrence of sea breezes. With this difficulty in mind, we have found that it is even more important to bring a particle counter to the measurement site. Namely, if the wind measurements are showing that the ships should be affecting the air quality at the measurement site, but the particle counter is not measuring any detectable plume for any ships, this indicates that the winds from the ships are not reaching the measurement site. Author's changes in manuscript 2. We have added a recommendation in the section "5 Recommendation and concluding remarks": "Before performing the measurements with the new method, it is important to investigate the meteorological situations at the current measurement site. For example, during sea breezes, local

wind measurements could indicate that shipping lane emissions should reach the measurement station, whereas in reality they might not. Care should be taken to account for these periods when the meteorological data will give erroneous results. However, these meteorological phenomena do not take place all the time, hence these specific meteorological conditions will not disqualify any chosen measurement site with the current proposed method. Again, these uncertain wind conditions make it very important to bring a particle counter to register shipping plumes. If the particle counter does not register any ship plumes during a selected time period, this indicates that winds from the ships are not reaching the measurement station, despite that the local wind measurements are suggesting otherwise."

Referee comment 3.

The paper is already very short. So why not including all the supplemental figures, tables, text, and references in the main paper? This way the paper will be much easier to read without having to refer to multiple documents.

Author's response 3.

We do not have a strong opinion against this suggestion. The placement of some information in the supplement was mainly an attempt to make the paper short and concise. But we agree with the point provided by the referee that it will not be practical for readers to look up information in the supplement. And if readers are interested, it can be a good idea to have all information easily accessible, since the manuscript is rather short at the moment. Re-considering the division between the manuscript and the supplement, we think that all information suits well in the original manuscript, except for the measurements of diffusion losses (Table S2 and Figure S2) due to its technical character. However, this is also such a short note that if the rest of the supplement is merged into the manuscript, it can be moved too, whereby we remove the supplement document entirely.

Author's changes in manuscript 3.

We have merged the supplementary information into the manuscript. The sections on measurement setup, meteorological parameters, and the characterisation of losses in the dryers is moved to "2. Instrumentation set-up and experimental site" and the section on log-normal parameters is moved to the section "4.2. Results of plume contribution calculations".

Referee comment 4.

The authors can compare their aerosol size distribution as a function of plume age to those reported by Aliabadi et al. 2015.

Author's response 4.

We have included the suggested scientific article in the section "1 Introduction", together with references to several other studies of size resolved particle number emissions from ships. Therefore, we have not included a specific comparison with the suggested size distribution, but rather given a broader background to the field to the reader. For comparing size distributions in detail, one has to consider the various differences between the many existing studies, which is relevant and possible to do, but outside of the main message of our manuscript. We are mainly interested in reporting the contribution to Falsterbo. But for future investigations the size distribution during longer transport times, it will be of value to compare with the suggested paper. Author's changes in manuscript 4. See "Author's changes in manuscript 2."

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