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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ship plumes in the Baltic Sea Sulphur Emission Control Area: Chemical characterization and contribution to coastal aerosol concentrations" *by* Stina Ausmeel et al.

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The study evaluates the properties of ship-related aerosols and the contribution of a major shipping lane to the coastal particulate matter and nitrogen dioxide based on measurements in individual ship plumes during one summer and one winter season at a site in Southern Sweden. The method of ship plume identification is based on a previous publication of Ausmeel et al. (Atmos. Meas. Tech., 12, 4479–4493, 2019) and has been shown to give a robust estimate of the ship contribution to the ambient concentrations downwind of a shipping lane. In addition, the ageing of ship aerosols is investigated in an oxidation flow reactor (OFR), which occasionally showed an increase

in sulfate and organic matter upon photochemical processing in air associated with the ship plumes. The paper gives a brief overview of the results obtained in the field study and the OFR study, and is generally well written. The Introduction is very brief and does not give a comprehensive review of literature on the topic of measurements and chemical characterization of ship emissions, see my specific points (2+3) below.

The study has been performed in the context of the new sulphur regulation that requires fuel sulphur content equal to or less than 0.1% by mass in 2015. It should be made clearer in the discussion of the results how the paper complements previous measurements or modelling of the ship contribution to ambient air concentrations before/after the new sulphur regulation. The new regulation has led to changes in the type of fuel used or to installation of scrubbers: I think the Introduction should inform more about the changes of the ship operation (fuels, engine operation, scrubbers, etc.) and their expected impact on exhaust composition, in response to the new sulfur regulation. The paper should also discuss whether the finding of relatively low contribution from shipping to coastal particle phase concentrations might suggest that the taken measures have not only affected the sulphur components but also other constituents of the ship exhaust.

The distance of 10 km, which corresponds approximately to a plume ageing time of 30 min, seems long, even if it is considerable shorter than in a previous similar study by Kivekäs et al. (2014). At this distance, the particle number concentrations in the plume are already approaching the particle number concentrations in background air. Due to this distance, there is a chance that ship plumes from somewhat further away, like 20 km, are already further diluted and might contribute either to the background or to the plume signal of a "detected" ship. It should be explained how the method deals with other ships in the source corridor and the possibility of crossing or coinciding/indistinguishable ship plumes.

The method relies entirely on the accuracy and time resolution of the N measurements of the SMPS or particle counter. Kivekäs et al. (2014) apply a criterion for plume

detection that the excess N has to be larger than 500 cm-3 for detection of a ship plume (one hour plume ageing). Why is no such criterion used here? Meandering of the plume would also cause a fluctuation of N measured, which may lead to obscured plume detection. How did you deal with rapid changes of the background N?

The study does not discuss the impact of meteorological conditions and atmospheric stability on the plume detection. A higher and well-mixed boundary layer allows more vertical mixing of the plume and would lead to lower particle numbers in the plume that can be measured at the site. There are also some effects due to the location of the measurement site at the interface between the marine boundary layer and the boundary layer over land. Sensible heat flux associated with solar heating of the land surface can lead to the growth of a thermal internal boundary layer (ITBL) at the shoreline. Stable onshore wind flow advected over a cold water surface during the daytime, when passing the shoreline can be eroded by the ITBL, that can intercept an elevated ship plume and fumigate it rapidly to the ground (Lyons and Cole, 1973; van Dop et al., 1979; Hanna et al., 1984; Nazir et al., 2005).

Aromatic VOC might also be good tracers for the ship plume. Toluene levels measured by PTR-TOF have been used to detect contamination by ship exhaust in a ship-borne measurement campaign (Chang et al., 2011). Were VOC measured at the site or is it considered to complement future campaigns with PTR?

Murphy et al. (2009) reported a study with simultaneous shipboard and airborne measurements of the chemical composition and water-uptake of particulate ship emissions. One important finding of that study was that the in-plume organic-to-sulfate mass ratio did not change with increased plume ageing, indicating that the ship-originated particulate phase is not volatile enough to repartition back to the gas phase as the plume dilutes further. Please provide OC:$SO4_{dry}$ ratios measured in the ship plumes at Fallsterbo (and compare to values in the literature) as they could be helpful in deriving emission factors of OC for the ship fleet in the Baltic Sea. The ratio could also give valuable information about the ships with high sulfate formation in the OFR, obviously

not complying with the new sulfur regulation.

Specific Comments:

1.) P. 1 line 18: Please give percentage fraction of plumes that did not result in measurable secondary PM.

2.) P. 2 line 48 - 50: It would be good to add a review of PM emission factors obtained by the CO2 method, which is probably the most accurate method to infer real-world emissions, and compare to the PM emission factors obtained from testbed experiments.

3.) P. 2 line 50 - 51: Please explain: “It is difficult to simulate atmospheric dilution in testbed experiments, which has large effects on nucleated nanoparticles”. Please explain the effects on nucleated nanoparticles in more details and add literature references. Does “atmospheric dilution” refer to the fast cooling and expansion of the exhaust plume at stack exit or the dilution due to atmospheric turbulence?

4.) P. 6 line 216 - 220: The explanation of the differences of PM0.15 in ship plumes to the study by Kivekäs et al. (2014) is somewhat speculative. What about the influence of different atmospheric chemistry regime or seasonal differences? The statement “these effects are likely cancelling each other out” is too strong. Simulations and observations of the particle number concentrations in ship plumes reported by Tian et al. (2014, figure 3 therein) show that after rapid dilution in the first minutes, total particle number concentration change only slowly between 30 and 60 minutes. This means that the effect of longer dilution period would be small. On the other hand, the ship contribution determined by Kivekäs et al. covers a larger source corridor, a fact that is not discussed here.

5.) P. 7 line 267 - 269: The average BC fraction of 2% of the total PM0.5 mass is very low compared to previous studies of ship exhaust, which may be explained in part by different fuels, operating conditions or the use of scrubbers. For a better understanding of this result, please provide the BC fraction, if only the plumes with detectable eBC

increase were included in the analysis.

6.) P. 9 line 339-340: Please discuss in more detail the (only) moderate increase of nitrate and ammonium in the OFR. Formation of ammonium nitrate requires the presence of sufficient ammonia, which may be the limiting factor during the time when the measurements were made. Thus, nitrate and ammonia could be higher during the spring season with more agricultural activity. The gas-phase/particle partitioning is also very sensitive to changes in temperature and relative humidity inside the reactor compared to ambient air.

7.) P. 9 line 351-353: How likely is it that heterogeneous oxidation of organic aerosols happens in the real plume ageing where the environment is much less oxidative?

Technical Corrections:

P. 4 line 143: “Chemical composition” would be better here.

Figure 1: Please add a panel with wind direction and wind speed below the current plots.

Figure 3: The orange line for ammonium is hardly visible.

Table 1: Please give the number of plumes in summer and winter somewhere in this table. It would be better to replace “average plume concentration” by “Delta plume” since the column gives the excess concentration due to the ship plume and not the in-plume concentration.

References:

Chang, R. Y.-W., Leck, C., Graus, M., Müller, M., Paatero, J., Burkhart, J. F., Stohl, A., Orr, L. H., Hayden, K., Li, S.-M., Hansel, A., Tjernström, M., Leaitch, W. R., and Abbatt, J. P. D.: Aerosol composition and sources in the central Arctic Ocean during ASCOS, Atmos. Chem. Phys., 11, 10619–10636, https://doi.org/10.5194/acp-11-10619-2011, 2011.

Hanna, S. R., Paine, R. J., and Schulman, L. L.: Overwater dispersion in coastal regions, Boundary-Layer Meteorol., 30(1-4), 389−411, https://doi.org/10.1007/BF00121963, 1984.

Lyons, W. A. and Cole, H. S.: Fumigation and plume trapping on the shores of Lake Michigan during stable onshore flow, J. Appl. Meteor., 12, 494−510, 1973.

Murphy, S. M., Agrawal, H., Sorooshian, A., Padró, L. T., Gates, H., Hersey, S., Welch, W., Jung, H., Miller, J., and Cocker III, D. R.: Comprehensive simultaneous shipboard and airborne characterization of exhaust from a modern container ship at sea, Environ. Sci. Technol., 43, 4626−4640, 2009.

Nazir, M., Khan, F. I., and Husain, T.: Revised estimates for continuous shoreline fumigation: a PDF approach, Journal of Hazardous Materials 118, 53−65, doi:10.1016/j.jhazmat.2004.10.008, 2005.

Tian, J., Riemer, N., West, M., Pfaffenberger, L., Schlager, H., and Petzold, A.: Modeling the evolution of aerosol particles in a ship plume using PartMC-MOSAIC, Atmos. Chem. Phys., 14, 5327–5347, https://doi.org/10.5194/acp-14-5327-2014, 2014.

van Dop, H., Steenkist, R., and Nieuwstadt, F. T. M.: Revised estimates for continuous shoreline fumigation, J. Appl. Meteorol. 18, 133–137, 1979.

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