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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effects of ship emissions on air quality in the Baltic Sea region simulated with three different chemistry transport models" *by* Matthias Karl et al.

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We thank Referee #1 for positive evaluation of the manuscript. Following the Reviewer's remarks, the evaluation of total air concentrations and ship-related concentration contributions has been restructured in the revised manuscript. We have addressed the issues brought up below, with specific pointers to changed parts of the manuscript.

1. a) Several major aspects need to be addressed, in my opinion, prior to publication: 1) the MS should be shortened, as some sections are repetitive and feel like a report;

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Discussion paper

Reply: The manuscript has been shortened by significantly reducing the text of the Introduction, shortening the comparison with measurements, and removing the section on nitrogen deposition results. In particular, Sect. 3.1 ("Comparison to observations") was completely restructured to avoid the report-style text blocks.

1. b) 2) I would suggest to change the Figures and add correlation plots to show more of the data used for model validation, which are now not evident in the MS;

Reply: Figures showing time series plots of NO2 at coastal sites have been included in the new Appendix B. Time series plots of O3 at coastal sites are included in the SI, while all other time plots were removed. A new figure showing the spatial correlation (scatterplots with individual station points) as additional analysis of the model performance was included. We have kept the overview boxplots (Figures 3 - 6 in the original manuscript) because they give a compact overview of the three CTMs within one plot.

1. c) 3) model validation of the shipping contributions, was it carried out? it looks like no validation was performed, and this would need to be added, even if briefly;

Reply: This is not true. Although a direct comparison of the ship contribution was not carried out due to the methodological discrepancies with ship plume measurements (more explanations below), an evaluation method for the ship contribution had already been included in the original manuscript (Sect. 2.3.2 "Significance of the ship contribution"). Following the comments of this Reviewer, we have revised the previous method for testing the significance of the ship contribution. We present the evaluation in the new Sect. 3.2 "Evaluation of ship-related concentration contributions" of the revised manuscript.

1. d) 4) if possible, add recommendations for users as to which model performs better under which scenarios.

Reply: We have added a new section 3.3.5 on recommendations for model users in

the revised manuscript.

2. Specific comments: - p1, l12-13: contradiction, is the performance of the models similar or does it differ for PM2.5 in summer?

Reply: The sentence has been removed in the Abstract and also in the Conclusions (p. 24, line 1).

3. Introduction: can be shortened, specifically the paras dealing with CLs, literature review and SHEBA.

Reply: Introduction has been shortened in the revised manuscript.

4. p5, l23: are these total pollutant concentrations, or the ship-sourced fraction? This should be clarified throughout the text

Reply: This has been clarified here and at the other relevant places in the text. The title of Sect. 2.3.1 has been changed to "Evaluation method for the total air pollutant concentrations". The title of Sect. 3.1 has been changed to "Statistical evaluation of air pollutant concentrations". The significance of the ship contribution to total NO2 concentrations at monitoring stations has been evaluated in Sect. 3.1.5. This section has been renumbered as new Sect. 3.2 with title: "Evaluation of ship-related concentration contributions".

5. p6, l11-14: what about the non-linearity of O3? This approach (removing a source completely) has been seen to have higher uncertainty than if the source is only partially removed (e.g., decreasing its contribution by a given %), given that complete removal of the source doesn't take into account the non-linearity of certain species (e.g., O3). Please discuss how this may have affected the results.

Reply: The procedure of deducing the ship contribution from one run including all emissions and one run without the emissions from shipping (zero-out method), which is used here, assumes linearity. The perturbation of the ship emissions, for example

reduction by 20 %, as suggested by the Reviewer might be more careful with respect to the non-linearity of the involved photochemistry. However, our goal was to derive the full impact of shipping, while perturbing is mainly used to investigate the response to small changes (e.g. 20 %) of sectoral emissions. The assumption of linearity is reasonable. Moreover, the influence of shipping on ozone at coastal sites was found to be small. The following was added on p. 6 line 14:

"Previous calculations have shown that the assumption of linearity, by adding the contributions from different emission sources, is reasonable for ozone and other pollutants, and that the associated error is within a few percent (Jonson et al., 2018a; Karl et al., 2019)."

6. - p8, l28: the use of monthly averaged gridded emissions is indeed a major difference between the models; wouldn't it have an impact also on the underestimation of titration, as described above for the spatial resolution (p4, l5-10)?

Reply: The use of monthly averaged ship emissions in the EMEP model, versus daily emissions, is explained in Sect. 2.2.5. Initial tests had been performed with the EMEP model using both daily aggregated and monthly aggregated ship emissions of STEAM (from FMI). The differences of the model results were small, including for ozone. The statistical evaluation with ozone concentration measurements showed hardly any differences between the two setups. In particular, differences at coastal sites were of the order of +/- 0.01 ppb or less.

7. The paper is very well referenced, in general.

Reply: Thank you.

8. p11, l11: the model results were validated for total pollutant concentrations(e.g., against Airbase observation), and for ship-sourced pollutant concentrations (in this case, against what?)? Or only for total concentrations? PLease clarify.

Reply: We clarify that Sect. 2.3.1 (“Evaluation method for the total air pollutant concentrations”) describes the method for evaluation of the total pollutant concentration and Sect. 2.3.2 (“Significance of the ship contribution”) describes the method for evaluation of the ship contribution to the observed total concentration. The test of the significance of the ship influence at the monitoring stations can be regarded as an evaluation of the ship-related concentration, since it demonstrates how much the prediction of observed concentrations improves when shipping emissions are included in the simulation. The significance test has been repeated and the results of the evaluation are presented in the new Sect. 3.2 (“Evaluation of ship-related concentration contributions”). Including ship emissions improves the agreement between modelled and measured total NO2 daily mean concentrations at about 50 %) of the stations.

9. p11, Fig S1: it is not practical for the reader to start the Results section with Figs which are in Supporting Info. Maybe the authors could add the 4 time series at the bottom of Fig 3, for example?

Reply: We assume that this point refers to page 13 (first reference to Fig. S1). Section 3.1 has been restructured in accordance with the next points of this Reviewer. The time series plots for the selected two rural and two urban sites were removed from the SI (i.e. Fig. S1 - S4). The comparison has been merged with the analysis of the ship contribution at coastal stations. We therefore discuss the stations at shoreline and harbour cities in new Sect. 3.2 “Evaluation of ship-related concentration contributions” (Sect. 3.1.5 in the original manuscript). Time series plots of NO2 at coastal sites are now in the Appendix B. Time series plots of O3 at coastal sites are now in the new Fig. S1 of the SI.

10. p11, l27: stations were grouped as rural and urban background, why? It would be more useful to see the individual points, instead of the averages, to have additional detail.

Reply: The AirBase observation database has two main categories: rural background

stations and urban background stations. Traffic and industrial sites were excluded since regional CTM systems applied in this study do not handle local scale dispersion (see p. 11, lines 29 - 32). Statistics for temporal correlations between modelled and observed total concentrations at individual stations are given in the SI (Tables S3 - S6). However, we now included the spatial correlation (scatterplots with individual station points) as additional analysis of the model performance.

11. p13, l12: why were these 2 rural stations selected? Please clarify the criteria, here and for other pollutants.

Reply: The time series plots for the selected two rural and two urban sites were removed from the SI (i.e. Fig. S1 - S4) because the choice of the stations was arbitrary and the discussion of the time series plots did not provide additional information to the model evaluation. Instead, a spatial correlation analysis was added, see next point.

12. also in this section, instead of selecting 4 sites, what about plotting all of them in a correlation plot, for example the summer and winter mean per site? This would be helpful because with the current boxplot it is not so easy to see whether there is under or overestimation.

Reply: As recommended by this Reviewer, we have added scatterplots for the analysis of the spatial correlation of the annual mean total pollutant concentration together with the seasonal averages in the new Figure 7 and discuss this in the new subsection 3.1.2 “Spatial correlation”. However, we prefer to keep the overview boxplots (Figures 3 - 6) because they give a compact overview and comparison of the statistics of the daily mean values from three CTMs within one plot.

13. subsections in section 3.1: their structure sounds a bit too much like a report, they are too similar (only changing the pollutant). Suggestion to redraft and shorten.

Reply: We followed the suggestion of this Reviewer to redraft and shorten Sect. 3.1.

Table 3 has been moved to the Supplement. Information on statistical indicators can be seen in the boxplot figures, Fig. 3-6 (R, NMB and RMSE) and in the SI Tables (Tables S3-S6). Therefore, we have removed this information from the text. Sect. 3.1 has been restructured into the following subsections: 3.1.1 Rural versus urban sites and 3.1.2 Spatial correlation.

14. section 3.1.3 (SO2); what is the reason for the poorer performance of models for SO2 at rural sites? There are larger differences between models, too. Please provide an explanation.

Reply: The poorer performance of the models at the rural sites is related to uncertainties of local residential heating emissions. The following has been added on p. 15, line 29 of the original manuscript:

"The weaker performance of the models for SO2 at the rural sites is related to uncertainties of local residential heating emissions, as the timing of use and the sulfur content of burned fuels are difficult to predict."

15. p19, l4: recommendation to add a short concluding section on the comparison between models? This could include recommendations for users as to which model to select depending of the input data available or the purpose of the study.

Reply: Much of the discrepancies between the CTMs depend on the model configuration: ship emission, land-based emission, meteorology, and boundary conditions. We have added a new section 3.3.5 ("Recommendations from the comparison between the CTM systems") where we briefly evaluate the three models in terms of input data requirements, required level of user experience and model performance based on experience from this comparison. In addition, we give recommendations for which type and purpose of the study each model is suited best.

16. section 3.3: are the modelling contributions of shipping emissions validated

in any way? A comparison with point locations could be carried out, based on literature review (even if the observational data correspond to different years, a qualitative comparison would still be necessary). Source apportionment studies should be used for this validation.

Reply: The evaluation of ship-related concentrations (of NO2) had been presented in Sect. 3.1.5 of the original manuscript. As mentioned in response to previous points of this Reviewer, we have revised the test of the significance of the ship contribution. We present the evaluation in the new Sect. 3.2 "Evaluation of ship-related concentration contributions". The evaluation of the ship contribution is only done for NO2 (daily means), since ship emissions are known to be a relevant contributor to ambient NO2 concentrations and NO2 is monitored at many stations in the coastal regions. In our view, it is not possible to compare the modelled ship-related concentration directly to measurements of the ship contribution at point locations. Usually, these point observations report the concentrations in the plume from a single ship (or a few ships), passing the site, in exceedance of the measured background value. In the models, emissions from all ships at sea within one hour and within a radius of up to 50 km upwind contribute to the ship signal at a point location. In particular for PM2.5 this can lead to large discrepancies between the two methods: during atmospheric transport of emitted pollutants, oxidation and condensation happens, leading to a high fraction of secondary aerosol to the PM2.5 signal, whereas point measurements mainly capture the contribution of primary particles to the PM2.5 signal. Performing source apportionment studies with the models is out of scope of the paper because the main goal is to quantify the effect of shipping emissions. Source apportionment would require tagging of all possible emission sectors that contribute to the total concentration at a point location. In addition, emissions from other sectors are associated with uncertainties, which might be even higher than that of shipping.

17. p19, l12: would it be possible to provide an average for coastal areas? Or a range? This is usually where most population is exposed. This would be very

useful for all the other pollutants as well.

Reply: The spatial averaged ship contribution in the coastal land areas was added in Table 5, Table 6 and Table S10. The information on the average for coastal areas was also included in the manuscript text and the abstract.

18. p21, l31: the ship-related EC concentrations are really low, were these data validated in any way? If not, please state clearly.

Reply: The relatively low values for the modelled ship-related EC concentrations appear to be justified based on a comparison with data measured at a shoreline location in southern Sweden, which report even lower contributions from ships. Despite the limitations mentioned in our reply to the previous point on evaluating modelled ship contributions, such a comparison gives some guidance regarding the plausibility of the modelled EC values. The following has been added to the manuscript (p.21, line 31):

"Measurements of the ship contribution to equivalent black carbon (eBC) concentrations at a shoreline location in southern Sweden (Falsterbo [55.3843 N, 12.8164 E] downwind of main shipping lanes, based on 113 individual plumes, reported a value of 0.0035 μg m-3 as average of the winter campaign in 2016 (Ausmeel et al., 2019). Wintertime average modelled ship-related EC at this location is factor 4 to 6 higher than the measured value (CMAQ: 0.0207 μg m-3; SILAM: 0.0144 μg m-3, EMEP/MSC-W: 0.0149 μg m-3). The discrepancy might arise from comparison with a different year than used in the model simulations. Another reason for the higher model values is that the CTMs consider all ships within a radius of 50 km upwind, whereas measurements considered individual ships passing by in a limited sea area."

19. section 3.6: the deposition section doesn't seem to fit in this MS, could it be included in the Karl et al. companion paper, instead? Otherwise, suggestion to remove it.

Reply: Section 3.7 ("Comparison of oxidized nitrogen deposition") and Figure 11 have

been removed.

20. p23, l25: the uncertainties of the models or atmospheric transport and transformation of pollutants were not addressed in the MS; please add this in the new section on recommendations and conclusions, or remove this phrase.

Reply: The sentence has been removed.

References:

Ausmeel, S., Eriksson, A., Ahlberg, E., and Kristensson, A: Methods for identifying aged ship plumes and estimating contribution to aerosol exposure downwind of shipping lanes, Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-445, in review, 2019.

Jonson, J., Gauss, M., Schulz, M., and Nyíri, A.: Emissions from international shipping, in: Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components, EMEP Status Report 1/2018, pp. 83-89, Norwegian Meteorological Institute, Oslo, Norway, 2018a.

Karl, M., Bieser, J., Geyer, B., Matthias, V., Jalkanen, J.-P., Johansson, L., and Fridell, E.: Impact of a nitrogen emission control area (NECA) on the future air quality and nitrogen deposition to seawater in the Baltic Sea region, Atmos. Chem. Phys., 19, 1721-1752, https://doi.org/10.5194/acp-19-1721-2019, 2019.

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