

Changes to manuscript ms-nr acp-2018-1317

Effects of ship emissions on air quality in the Baltic Sea region simulated with three different chemistry transport models

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Dear Dr Huan Liu,

We highly appreciate the reviews of our manuscript ms-nr acp-2018-1317 that we received from you and one anonymous referee. We have replied to their comments in the Open Discussion. We have addressed all specific comments in the revised manuscript as will be described below. We carefully considered the concerns expressed by the anonymous referee and in the Editor Comment in our revision of the manuscript.

Below follows: (1) the point-by-point replies to the two reviewers, (2) a list of relevant changes in the manuscript, and (3) the revised manuscript with changes highlighted.

Our responses to reviewers have been written in **blue font**.

Figure, table, section, and page numbers in the replies below refer to the original manuscript. The revised manuscript with changes highlighted has been sent along with this response.

Referee #1

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;

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;

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;

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.

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 PM_{2.5} in summer?

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.

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

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

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)."

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.

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)?.

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.

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.

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?

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.

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.

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.

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.

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.

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.

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.

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 PM_{2.5} 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 PM_{2.5} signal, whereas point measurements mainly capture the contribution of primary particles to the PM_{2.5} 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.

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.

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 $\mu\text{g m}^{-3}$ 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 $\mu\text{g m}^{-3}$; SILAM: 0.0144 $\mu\text{g m}^{-3}$, EMEP/MSC-W: 0.0149 $\mu\text{g m}^{-3}$). 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.”

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.

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.

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

The sentence has been removed.

Editor Comment

1. The CMAQ version is too old to include advanced SOA mechanisms. Wildfire emissions were not included in emission inventory. All of above could be the reasons for low estimation on summer SOA. These disadvantages should be fixed or at least discussed.

The aerosol scheme AERO5 was applied in the CMAQ model, considering the SOA formation pathways based on traditional two-product representation, including reaction of volatile organic compounds to give non-volatile products, oxidative ageing of primary organic aerosol, acid-catalysed enhancement of SOA mass, oligomerization reactions and in-cloud aqueous-phase oxidation. In CMAQ v5.2, the aerosol scheme AERO6 with multi-generational aging chemistry was introduced. This version had not been available at the time when the CMAQ simulations for this study were performed (2016). Primary organic aerosol (POA) previously treated as non-volatile and non-reactive, can evaporate, oxidize, and re-condense to form SOA, which is known as multi-generational aging of primary organic aerosol (Robinson et al., 2007). The multi-generational aging chemistry for the semi-volatile POA configuration introduced in CMAQ v5.2 is derived from the approach of Donahue et al. (2012) which takes into account the functionalization and fragmentation of organic vapours upon oxidation. However, atmospheric SOA processes are still not fully understood and models have difficulties with prediction of SOA (Jimenez et al., 2009). A discussion of the disadvantages of the used CMAQ version has been added in section 3.4:

“The SOA formation mechanism in the applied version of CMAQ (i.e. v5.0.1) is probably not adequate for reproducing the summertime aerosol. Primary organic aerosol (POA), SOA and organic vapours in the atmosphere should be considered a dynamic system that constantly evolves due to multi-generation oxidation (Robinson et al., 2007). We note that multi-generational aging chemistry for the semi-volatile POA was introduced in CMAQ v5.2, based on the approach of Donahue et al. (2012) which considers the functionalization and fragmentation of organic vapours upon oxidation. In addition, wildfire emissions have not been considered in the simulation with CMAQ. Wildfires emit large quantities of organic material and are associated with high biogenic VOC emissions due to high temperature, leading to increased SOA formation (Lee et al., 2008).”

Donahue, N. M., Kroll, J. H., Pandis, S. N., and Robinson, A. L.: A two-dimensional volatility basis set - Part 2: Diagnostics of organic aerosol evolution, *Atmos. Chem. Phys.*, 12(2), 615–634, doi:10.5194/acp-12-615-2012, 2012.

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevôt, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S.,

Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of organic aerosols in the atmosphere, *Science* 326, 1525–1529, doi: 10.1126/science.1180353, 2009.

Lee, S., Kim, H. K., Yan, B., Cobb, C. E., Hennigan, C., Nichols, S., Chamber, M., Edgerton, E. S., Jansen, J. J., Hu, Y., Zheng, M., Weber, R. J., and Russell, A. G.: Diagnosis of aged prescribed burning plumes impacting an urban area, *Environ. Sci. Technol.*, 42(5), 1438–1444 doi:10.1021/es7023059, 2008.

Robinson, A. L., Donahue, N. M., Shrivastava, M. K., Weitkamp, E. A., Sage, A. M., Grieshop, A. P., Lane, T. E. and Pierce, J. R., and Pandis, S. N.: Rethinking Organic Aerosols: semivolatile emissions and photochemical aging, *Science* 315, 1259–1262, 2007.

2. Authors should add the model validation for meteorology parameters.

Reviewer #1 has already asked to shorten the manuscript. Therefore, we refrain from adding a detailed evaluation of the meteorological fields used in each of the three CTMs. Regarding the evaluation of WRF used in the SILAM simulation we refer to the study by Kryza et al. (2017) where a WRF setup with similar configuration and spatial resolution has been evaluated with station measurements in Poland. For the EMEP model we refer to the evaluation of the ECMWF weather forecast from cycle Cy40r1 as summarized in Haiden et al. (2014). The meteorological fields from the COSMO-CLM model that were used in the CMAQ simulation have been evaluated with respect to frequency and amount of precipitation in Karl et al. (2019). We include an evaluation of the 2 m air temperature (T2) and the wind speed at 10 m (WS10) of the 0.025 degree COSMO-CLM data in the southern part of the Baltic Sea (BS) domain.

Temperature was compared against gridded observational data from E-OBS v.16 (Cornes et al., 2018) of the European Climate Assessment & Dataset (ECA&D). Wind speed was compared against observational data from MiKlip DecReg of the German Weather Service (DWD). Monthly T2 in Denmark and southern Sweden (Fig. C1) was underestimated in winter (bias smaller than -1.4 K) and overestimated in summer. The warm bias in summer was higher in Sweden (+1.4 K) than in Denmark (+0.4 K). In contrast to southern Sweden, the winter T2 was overestimated in the more northern parts of Sweden (Fig. C2). The spatial correlation of T2 in the southern BS domain based on 3-daily averages was remarkably good ($R = 0.94$). Monthly WS10 was slightly overestimated in most parts of the southern BS domain (Fig. C3). The largest errors of wind speed occurred in Denmark and northern Poland during May and June.

The summary of the evaluation of meteorological variables has been added to section 2.2.2 (“Meteorology”) in the revised manuscript.

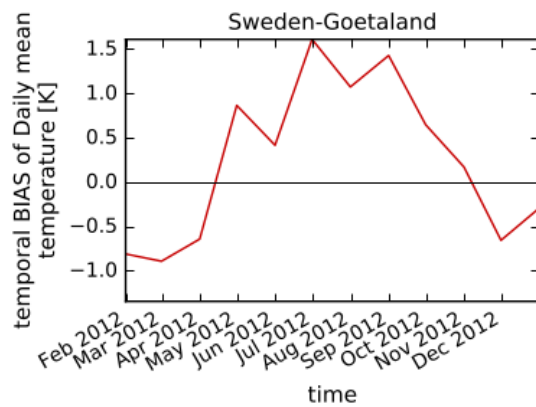
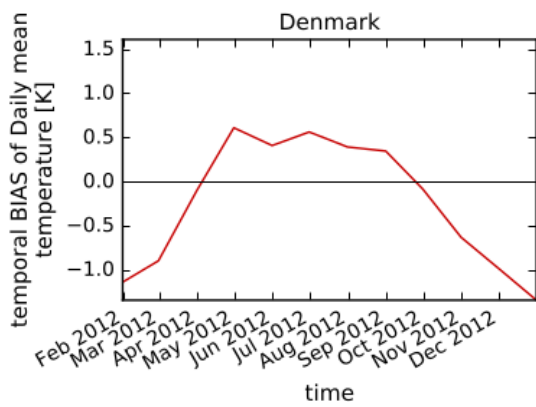
Cornes, R., van der Schrier, G., van den Besselaar, E. J. M., and Jones, P. D.: An ensemble version of the E-OBS temperature and precipitation datasets, *J. Geophys. Res. Atmos.*, 123, doi:10.1029/2017JD028200, 2018.

Haiden, T., Janousek, M., Bauer, P., Bidlot, J., Ferranti, L., Hewson, T., Prates, F., Richardson, D. S., and Vitart, F.: Evaluation of ECMWF forecasts, including 2013-2014 upgrades, ECMWF Technical Memorandum No. 742, December 2014, European Centre for Medium-Range Weather Forecasts, Reading, UK, 2014.

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.

Kryza, M., Walaszek, K., Ojrzynska, H., Szymanowski, M., Werner, M., and Dore, A. J.: High-resolution dynamical downscaling of ERA-Interim using the WRF regional climate model for the area of Poland. Part 1: Model configuration and statistical evaluation for the 1981-2010 period, *Pure Appl. Geophys.*, 174(2), 511-526, doi:10.1007/s00024-016-1272-5, 2017.

a)



b)

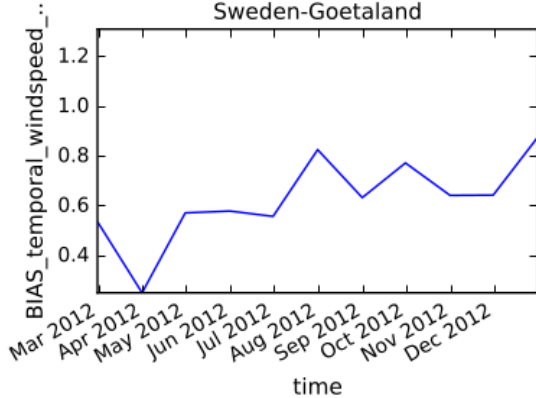
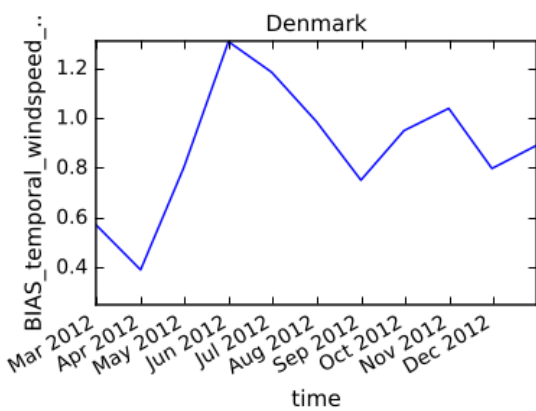


Figure C1: Evaluation of COSMO-CLM data: temporal bias a) of the monthly mean 2 m air temperature in Denmark (left) and southern Sweden (right); and b) of the monthly mean 10 m wind speed in Denmark (left) and southern Sweden (right).

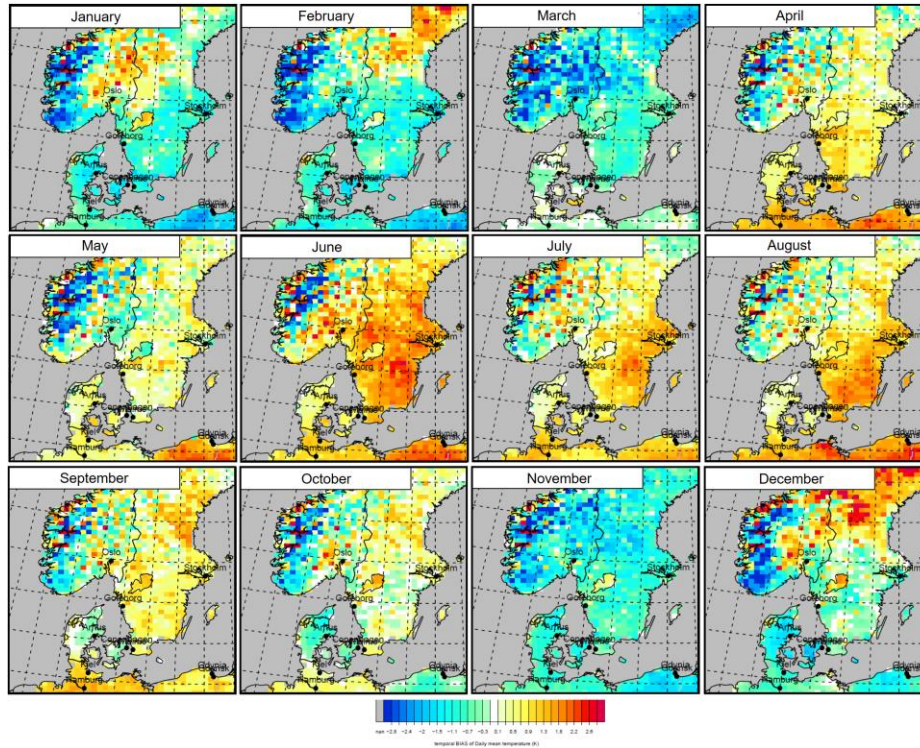


Figure C2: Evaluation of COSMO-CLM data: spatial distribution of the temporal bias of the monthly mean 2 m air temperature in the southern part of the Baltic Sea region.

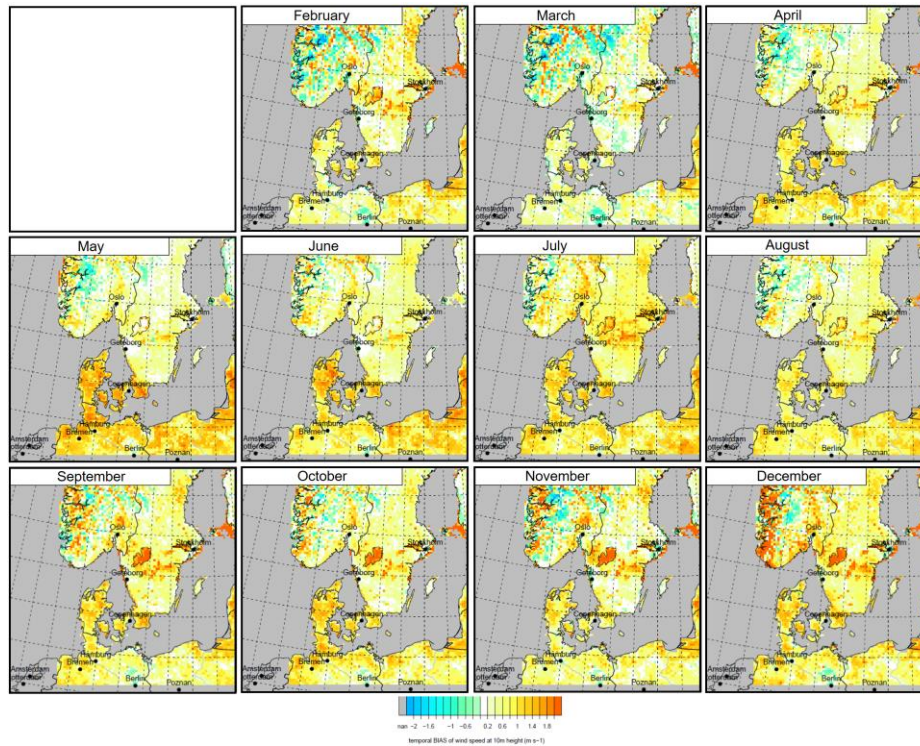


Figure C3: Evaluation of COSMO-CLM data: spatial distribution of the temporal bias of the monthly mean 10 m wind speed in the southern part of the Baltic Sea region. Missing observation data for January 2012.

3. Table 2, how do you get the “average fractions of the total emission in each vertical model layer”? This factor and its source need a very detailed description. Why the highest emission could reach 1000m in SILAM model? If this is true, the deposition process would be influence a lot.

The average fractions of total emissions in each vertical model layer are derived from the two STEAM datasets, one containing ship emissions below 36 m and one containing ship emissions above 36 m (p. 11, line 3). The dataset with ship emissions above 36 m was intended to represent the emissions of ships with a stack exit above 36 m above sea level. Ship emissions from the two datasets were distributed into the different vertical layers of the CTMs. This had been done differently for the three models because of the different model layer heights. Unfortunately, further analysis of the SILAM results revealed, that the STEAM data for ship emissions above 36 m had been injected in the model’s vertical layers between 36 m up to 1000 m height. We agree that this procedure was erroneous. Consequently, we have repeated the SILAM “base” run with ship emissions vertically distributed in the same kind as in the CMAQ model. New results from the SILAM run show higher ship contribution of NO₂, SO₂, and more ozone titration. Further, the statements about the effect of the vertical ship emission profile on ship-related EC concentrations of SILAM in Sect. 3.5 (“Comparison of elemental carbon related to ship emissions”) have been removed. Table 2 has been removed because it is now unnecessary.

4. The references and equations for NMB, R, RMSE and FAC2 should be added.

Definitions of these statistical indicators are now given in an Appendix A.

5. The last sentence in section 3.3.2 is not accurate. It should be “NO_x-limited regime in the model”.

The results from the new SILAM “base” run show a similar tendency for annual mean O₃ concentration changes due to shipping as the other two models. The referred sentence has therefore been removed.

List of relevant changes in the ms

Relevant text changes:

1. Introduction

As requested by Referee #1, the Introduction has been shortened.

2. Section 2 “Description of the CTM systems and setup of the model comparison”

Following the Comment from the Editor, we have added a brief evaluation of the meteorological fields used in each of the three CTMs in Sect. 2.2.2 (“Meteorology”) (p.9, line 18):

“The SILAM model is run with meteorological input from a simulation with the Weather Research and Forecast (WRF) model v3.7.1 using original resolutions of 4.0 km, 16.0 km, and 64.0 km, for inner, central and outer domains, respectively. WRF was driven with large scale meteorological forcing data taken from the ERA-Interim reanalysis of ECMWF (Dee et al. 2011). The high-resolution inner domain extended up to 2000 m height and was therefore less influenced by upper tropospheric dynamics of WRF. Kryza et al. (2017) using WRF in a similar configuration evaluated the WRF meteorological fields against station observations in Poland. The 2 m air temperature (T2) was underestimated in winter (bias smaller than -0.6 K) while temperature in the warm season was overestimated (bias up to +1.0 K). The largest errors for the 10 m wind speed (WS10) occurred in late summer and autumn and the largest errors for wind direction (WDIR) in spring and summer. The error of wind direction was very small in winter. Spatial distribution of meteorological variables obtained from WRF were in close agreement with the station measurements, but the model performance was found to be worse for the seashore and mountain areas than for other inland areas (Kryza et al., 2017).

High-resolution meteorological fields for CMAQ were obtained from the COSMO-CLM (Rockel et al., 2008) model version 5.0. The meteorological fields were converted to the extension, resolution and projection of the CMAQ nested grids, using an in-house modified version of MCIP. More details on the meteorological forcing data and the evaluation of precipitation can be found in Karl et al. (2019). Here we include an evaluation of T2 and WS10 in the southern part of the Baltic Sea region. Temperature was compared against gridded observational dataset E-OBS v.16 (Cornes et al., 2018). Wind speed was compared against observational data from MiKlip DecReg of the German Weather Service (DWD). Monthly mean T2 in Denmark and southern Sweden was underestimated in winter (bias smaller than -1.4 K) and overestimated in summer. The warm bias in summer was higher in Sweden (+1.4 K) than in Denmark (+0.4 K). The spatial correlation of T2 in the southern Baltic Sea region based on 3-daily averages was remarkably good. Monthly mean WS10 was slightly overestimated in most parts of the region. The largest errors of wind speed occurred in Denmark and northern Poland during May and June.

EMEP/MS-CW was driven by meteorological data from the Integrated Forecasting System (IFS) of the ECMWF, version IFS38r2, with t1279 resolution (about 0.16 degrees resolution) interpolated to 0.1 × 0.1 degrees. The ECMWF forecasting system of weather parameters is regularly validated by

comparing against European synoptic observation data available on the Global Telecommunication System (GTS). The evaluation of the weather forecast from cycle Cy40r1 is summarized as follows (Haiden et al., 2014). The frequency of light precipitation is overestimated, with a bias of 1.2–1.4 mm d⁻¹ (for precipitation amounts > 1mm d⁻¹). T2 has a negative night-time temperature bias over Europe in winter and early spring. For total cloudiness, bias and standard deviation are small in 2012. For WS10, the standard deviation is low and the night-time bias is very small.”

Sect. 2.3 (“Statistical analysis”) has been modified to make it clearer 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. As requested by the Editor, the statistical indicators NMB, R, RMSE and FAC2 are now defined in a new Appendix A.

3. Comparison to observations

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 in the original manuscript. This section has been renumbered as new Sect. 3.2 with title: “Evaluation of ship-related concentration contributions”.

We followed the suggestion of Referee #1 to redraft and shorten Sect. 3.1. Sect. 3.1 has been restructured into the following subsections: 3.1.1 “Rural versus urban sites” and 3.1.2 “Spatial correlation”.

Sect. 3.1.1 presents the statistical evaluation of the temporal variation of total air pollutant concentrations. The performance of the models to simulate air pollutant concentrations is compared and discussed separately for the group of rural stations and for the group of urban stations in order to highlight differences in the predictive capability of the models for rural versus urban sites.

The discussion of time series plots for the selected two rural and two urban sites were removed from the manuscript and the corresponding figures in the SI have been deleted (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. 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. 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.

As recommended by Referee #1, 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”.

In the new Sect. 3.2 “Evaluation of ship-related concentration contributions”, we discuss the stations at shoreline and harbour cities. Time series plots of NO₂ at coastal sites are now in the new Appendix B. Time series plots of O₃ at coastal sites are now in the new Fig. S1 of the SI.

Due to the new Sect. 3.2, the subsequent sections of the Results section have been renumbered.

4. Section 3.3 “Comparison of the spatial distribution of air quality indicators”

Following a comment from Referee #1, we have added the spatial averaged ship contribution in the coastal land areas in Table 3 and 4 (Table 5 and 6 of the original manuscript) and Table S10, in addition to that for the entire Baltic Sea region (which included also the area of the sea). The information on the average for coastal areas was also included in the manuscript text and the abstract.

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.

5. Section 3.4 “Comparison of the ship contribution in the three CTMs”

Following the comment of the Editor on the vertical ship emission distribution in the SILAM model, we have repeated the SILAM “base” run with ship emissions vertically distributed in the same kind as in the CMAQ model. The ship contribution calculated by the SILAM model to the ambient air pollutant has changed. Consequently, we have revised the text of Sect. 3.4 “Comparison of the ship contribution in the three CTMs” and the information in Table 3 (Table 5 of the original manuscript).

6. Section 3.6 “Comparison of elemental carbon related to ship emissions”

Following the Editor Comment, a discussion of the disadvantages of the used CMAQ version with respect to SOA formation has been added in Sect. 3.5 “Comparison of PM_{2.5} in summer and autumn” (p. 20, line 25):

“The SOA formation mechanism in the applied version of CMAQ (i.e. v5.0.1) is probably not adequate for reproducing the summertime aerosol. Primary organic aerosol (POA), SOA and organic vapours in the atmosphere should be considered a dynamic system that constantly evolves due to multi-generation oxidation (Robinson et al., 2007). We note that multi-generational aging chemistry for the semi-volatile POA was introduced in CMAQ v5.2, based on the approach of Donahue et al. (2012) which considers the functionalization and fragmentation of organic vapours upon oxidation. In addition, wildfire emissions have not been considered in the simulation with CMAQ. Wildfires emit large quantities of organic

material and are associated with high biogenic VOC emissions due to high temperature, leading to increased SOA formation (Lee et al., 2008).”

7. Section 3.6 “Comparison of elemental carbon related to ship emissions”

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. 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 $0.0035 \mu\text{g m}^{-3}$ 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: $0.0207 \mu\text{g m}^{-3}$; SILAM: $0.0144 \mu\text{g m}^{-3}$, EMEP/MSC-W: $0.0149 \mu\text{g m}^{-3}$). 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.”

Following the comment of the Editor on the vertical ship emission distribution in the SILAM model, we have repeated the SILAM “base” run with ship emissions vertically distributed in the same kind as in the CMAQ model. The discussion of the impact of vertical emission profile on ship-related EC concentration has therefore been removed.

7. Deposition of nitrogen

Sect. 3.6 “Comparison of oxidised nitrogen deposition” of the original manuscript has been removed because of the concern expressed by Referee #1.

8. Summary and Conclusions

Following the concern expressed by Referee #1, the sentence on p.23, line 25 (original manuscript) about the uncertainties of the models or atmospheric transport and transformation of pollutants has been deleted.

The conclusion on the evaluation of ship-related contribution to ambient levels of NO₂ and O₃ has been revised. The statements about nitrogen deposition (p.25, line 3-6) and vertical ship emission profile (p.25, line 11-14) have been removed.

Tables:

Table 2.

The table has been deleted.

Table 3.

The table has been moved to the Supplementary Information and is now Table S6.

Table 4.

The table has been renumbered as Table 2.

Table 5.

The table has been renumbered as Table 3.

Table 6.

The table has been renumbered as Table 4.

Figures:

Figure 7.

As recommended by Referee #1, 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.

Figure 7 of the original manuscript has been renumbered as Figure 8.

Figure 8.

The figure has been renumbered as Figure 9.

Figure 9.

The figure has been renumbered as Figure 10.

Figure 10.

The figure has been renumbered as Figure 11.

Figure 11.

The figure has been deleted.

Figure B1.

The new Figure B1 in the new Appendix B contains the time series plots of NO₂ concentrations at selected ship-influenced monitoring stations for the meteorological year 2012.

Effects of ship emissions on air quality in the Baltic Sea region simulated with three different chemistry transport models

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Abstract. The Baltic Sea is highly frequented shipping area with busy shipping lanes close to densely populated regions. Exhaust emissions from ship traffic into the atmosphere are not only enhancing air pollution, they also affect the Baltic Sea environment through acidification and eutrophication of marine waters and surrounding terrestrial ecosystems. As part of the European BONUS project SHEBA (Sustainable Shipping and Environment of the Baltic Sea Region), the transport, chemical transformation and fate of atmospheric pollutants in the Baltic Sea region was simulated with three regional chemistry transport models (CTM) systems, CMAQ, EMEP/MSC-W and SILAM with grid resolutions between 4 km and 11 km. The main goal was to quantify the effect that shipping emissions have on the regional air quality in the Baltic Sea region when the same shipping emissions dataset but different CTMs in their typical setups are used. The performance of these models and the shipping contribution to the results of the individual models was evaluated for sulphur dioxide (SO₂), nitrogen dioxide (NO₂) and ozone (O₃) and particulate matter (PM_{2.5}). Model results from the three CTMs for total air pollutant concentrations were compared to observations from rural and urban background stations of the AirBase monitoring network in the coastal areas of the Baltic Sea region. ~~The performance of the three CTM systems to predict pollutant concentrations is similar. However, observed~~ Observed PM_{2.5} in summer was underestimated strongly by CMAQ and to some extent by EMEP/MSC-W. Observed PM_{2.5} in winter was underestimated by SILAM. In autumn all models were in better agreement with observed PM_{2.5}. The spatial average of annual mean O₃ in the EMEP/MSC-W simulation ~~is 15–25~~ was ca. 20 % higher compared to the other two simulations, which is mainly the consequence of using a different set of boundary conditions for the European model domain. There are significant differences in the calculated ship contributions to the levels of air pollutants among the three models. ~~SILAM predicted a much~~ EMEP/MSC-W, with the coarsest grid, predicted weaker ozone depletion through NO emissions in the proximity of the main shipping routes than the other two models. ~~In the entire Baltic Sea region the~~ The average contribution of ships to PM_{2.5} levels in coastal land areas is in the range of ~~4.3–6.5~~ 3.1–5.7 % for the three CTMs. Differences in ship-related PM_{2.5} between the models are mainly attributed to differences in the schemes for inorganic aerosol formation. ~~Inspection of~~ Differences in the ship-related elemental carbon (EC) ~~revealed that assumptions about the vertical ship emission profile can affect the dispersion and abundance of ship-related pollutants in the near-ground atmosphere. The models are in agreement regarding the ship-related deposition of oxidised nitrogen, reporting a ship contribution in the range of 21–23 as~~

~~atmospheric input to the Baltic Sea. Results among the CTMs can be explained by differences in the meteorological conditions, atmospheric transport processes, and the applied wet scavenging parameterizations. Overall, results~~ from the present study show the sensitivity of the ship contribution to combined uncertainties of boundary conditions, meteorological data and aerosol formation and deposition schemes. This is an important step towards a more reliable evaluation of policy options regarding emission regulations for ship traffic and the planned introduction of a nitrogen emission control area (NECA) in the Baltic Sea and the North Sea in 2021.

1 Introduction

International shipping is important for the economic exchange in Europe: almost 90 % of the European Union (EU) import and export freight trade is seaborne. Compared to other modes of transport such as trucks and air freight, shipping is far more energy efficient per ton of cargo. The Baltic Sea is one of the most densely trafficked sea regions in the world. Roughly 407,500 ship crossings in the Baltic Sea were recorded in 2012 (HELCOM, 2014), including passenger, cargo, tanker and other ship types. Maritime transport of goods between main EU ports and ports located in the Baltic Sea has a share of 22 % (in 2016) of the total shipping tonnages within European seas (EUROSTAT, 2018).

Ship traffic is associated with exhaust emissions of a wide range of air pollutants, among them nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$), black carbon (BC), sulphur dioxide (SO_2), non-methane volatile organic compounds (NMVOC) and particulate matter, as well as greenhouse gases (mainly carbon dioxide, CO_2). ~~Primary and secondary particulate matter from ship exhaust has been associated with adverse health effects (e.g. Corbett et al., 2007).~~ The emitted amounts and size spectrum of particulate matter depends on the type of fuel and its sulphur content and the ship engine type (e.g. Fridell et al., 2008; Moldanová et al., 2009). ~~For regulatory purposes, particulate matter is divided into the size fractions and according to aerodynamic diameter, where covers all particles with~~ Particulate matter with aerodynamic diameter less than $10 \mu\text{m}$ and those (PM_{10}) and with diameter less than $2.5 \mu\text{m}$ ($\text{PM}_{2.5}$) from ship exhaust has been associated with adverse health effects (e.g. Corbett et al., 2007). A global model study by Sofiev et al. (2018b) demonstrated the health benefits from reducing the ship-related fine particulate matter by low-sulphur ship fuels in densely populated, major-trading nations.

~~Emissions from ships are transported in the atmosphere over several hundreds of kilometres (Endresen et al., 2003).~~ The atmospheric transformation of primary emitted gases from shipping is especially relevant for the formation of ozone (O_3) and the deposition of sulphur and nitrogen compounds distant from the ship lanes (Eyring et al., 2010). ~~Emissions from ships are transported in the atmosphere over several hundreds of kilometres (Endresen et al., 2003).~~ Exhaust emissions from ship traffic in the Baltic Sea has the potential to degrade air quality in the coastal areas (Jonson et al., 2015) and to significantly affect the Baltic Sea environment through acidification and eutrophication of marine waters and surrounding terrestrial ecosystems (HELCOM, 2009; Bartnicki et al., 2011; Hunter et al., 2011; Raudsepp et al., 2013; Neumann et al., 2018a, b, c; Neumann et al., 2018). Acidification is a major challenge in the Baltic Sea region today where the critical load (CL) for acidification is exceeded especially in the southern part (Tsyro et al., 2018). ~~The CL for acid deposition is the value that must not be exceeded in order to maintain the soil's capacity to neutralise the excess acid. Between 2000 and 2016, the percentage of ecosystem areas in Europe~~

where CL (acidification) was exceeded decreased from 16 % to 6 % (Tsyro et al., 2018), partially as a result of stricter sulphur regulations for ships. Despite the considerable improvement concerning critical loads with respect to acidification, there are still regions in the Baltic Sea catchment, for which further reductions in acidification are desirable (Hettelingh et al., 2017). Atmospheric deposition of nitrogen compounds play a role in the eutrophication of the coastal marine environment (e.g., Paerl, 1995) and threaten biodiversity in forests, semi-natural vegetation, and freshwater catchments through excessive nitrogen input (Cofala et al., 2007). Even though exceedances of CL for eutrophication has decreased over the past decades, critical loads are still exceeded in about 65 % of the European ecosystems (Tsyro et al., 2018).

Air pollution from ships is increasingly controlled worldwide by the International Maritime Organization (IMO) through the Marine Pollution Convention (MARPOL) Annex VI - Regulations for the Prevention of Air Pollution from Ships (IMO, 2008). The Baltic Sea has been a sulphur emission control area (SECA) since May 2006, with stepwise reductions of the sulphur content in ship fuels; from 2015 onwards the sulphur content of any fuel oil used on-board ships within the Baltic Sea has to be 0.1 % or less (van Aardenne et al., 2013). ~~The MARPOL regulations on emissions of (TIER) are defined as function of year of installation and vessel speed. The TIER I standard was introduced in 2000 and is up to 10 % stricter than for ships built before 2000. TIER II was implemented in 2011 applying 15 % stricter standards than TIER I.~~ The effect of regulations of nitrogen emissions have been small until now, as these are only enforced for the new built ships. A nitrogen emission control area (NECA) for the Baltic Sea, North Sea and English Channel will become effective in 2021, ~~in which but only~~ new built ships have to comply with ~~TIER III, approximately 75 % stricter than TIER II. the new regulation~~ The consequences of establishing the NECA on the future air quality in the Baltic Sea region are investigated in the companion paper by ~~Karl et al. (2018)~~ [Karl et al. \(2019\)](#).

The transport, chemical transformation and fate of atmospheric pollutants in the Baltic Sea region can be simulated with 3-D chemistry transport model (CTM) systems. Previous air quality model studies related to shipping in the North Sea and Baltic Sea (Matthias et al., 2010; Hongisto, 2014; Jonson et al., 2015, 2018b; Matthias et al., 2016; Antturi et al., 2016; Claremar et al., 2017) used CTM systems to investigate the effect of implementation of MARPOL regulations on sulphur emissions by ships, the effect of establishing the NECA and other ship emission control scenarios. The studies quantified the contributions from shipping to the total air concentrations, deposition of nitrogen and sulphur, as well as air quality and health indicators.

Jonson et al. (2018b) studied the effects of shipping on the global scale, including the effects of shipping in the Baltic Sea and the North Sea, compared to total anthropogenic emissions in a global CTM with 0.5×0.5 degrees resolution. They found that a significant fraction, ranging from 5 % to more than 10 %, of the $PM_{2.5}$ and the depositions of nitrogen of anthropogenic origin in bordering countries can be attributed to ship emissions in the two sea areas. ~~For the deposition of sulphur, ship emission contributions are markedly lower as a result of the implementation of a stricter SECA (from 2015 onwards). For ground level contributions were found to vary depending on country and the choice of ozone metric ranging from negative contributions for annual average ozone to more than 10 % of the summertime phytotoxic ozone dose (POD1) for forests in some countries in the Baltic Sea region.~~

On the regional scale, Jonson et al. (2015) using the EMEP/MSC-W model (Simpson et al., 2012) with a horizontal resolution of $14 \times 14 \text{ km}^2$ assessed the effect of reduced sulphur content (2015 value of 0.1 %) and regulation of NECAs on the air

quality, deposition of nitrogen and related impacts on human health in the Baltic and North seas. Matthias et al. (2016) used the Community Multiscale Air Quality (CMAQ) model v4.7.1 (Byun and Schere, 2006) with a horizontal resolution of $24 \times 24 \text{ km}^2$ to investigate the effects of different future developments of shipping emissions in the North Sea area on air quality in the North Sea region. Antturi et al. (2016) used the SILAM (Sofiev et al., 2015) CTM system with spatial resolution of ca. $8 \times 8 \text{ km}^2$ in a cost-benefit analysis of the sulphur reduction policy in the Baltic Sea, but did not investigate the effects of shipping emissions on ozone concentration or nitrogen deposition. The study by Claremar et al. (2017) used the EMEP/MSC-W model with a much coarser resolution ($50 \times 50 \text{ km}^2$) ~~and gridded shipping emissions from the ENTEC inventory (Jonson et al., 2009) for 2011 and from TNO-MACC-III (Gauss et al., 2015) for 2013.~~ They find highest contribution of international shipping in the Baltic Sea and North Sea to ambient levels of air pollutants near large harbour cities and along the main shipping lanes; ~~with contributions to ambient levels of nitrogen dioxide (NO_2), NO_x , and of about 80 %, 80 %, 13 %, and 20 %, respectively (Claremar et al., 2017).~~

The use of relatively coarse model grids (coarser than 10-km resolution) in some of the previous CTM simulations raises concerns about non-linear chemical effects, particularly for O_3 , since a high source strength from shipping in the proximity to large land-based emissions (inside the same grid cell of the model) often results in very high levels of NO_x and excessive ozone titration (Jonson et al., 2009). Moreover, shipping releases large amounts of NO_x from a moving point source within the relatively clean maritime atmosphere. In regional CTMs, these NO_x emissions are diluted into large grid volumes, which can lead to a systematic overestimation of the ozone production and artificially increases the lifetime of NO_x (Von Glasow et al., 2003; Song et al., 2003; Vinken et al., 2011).

~~In the Baltic Sea area, movements of ships are gathered in the regional HELCOM (Baltic Marine Environment Protection Commission – Helsinki Commission) Automatic Identification System (AIS) network and database (-). Not all previous air quality studies in the Baltic Sea region used AIS ship position data, some use long averaging intervals for shipping emissions. For the North Sea region, Matthias et al. (2016) designed emission control scenarios based on a detailed emission inventory built upon AIS ship positions and a detailed ship characteristics database (Aulinger et al., 2016). The studies by Antturi et al. (2016) and Jonson et al. (2015) deployed the Ship Traffic Emission Assessment Model (STEAM; Jalkanen et al., 2009, 2012) which combines the AIS-based information with the detailed technical knowledge of the individual ships.~~

Despite previous model based work on the effects of shipping on air quality and human health in the Baltic Sea region, there is a need for more localised studies building on a much higher level of details, i.e. concerning shipping activity, for the quantification of regional ship-related air pollution. Knowledge on air quality impacts of shipping with a finer spatial resolution than in previous model studies is required for the identification of best suited sustainable development options for the shipping sector, especially if a varying suite of competing environmental and economic drivers is to be considered in different sub-regions.

With the goal to support EU policies on environmental and economic aspect of the shipping sector the BONUS project SHEBA (Sustainable Shipping and Environment of the Baltic Sea Region;-) was established in 2015. The overarching aim of BONUS SHEBA was an integrated and in-depth analysis of the ecological, economic and social impacts of shipping in the Baltic Sea. ~~The project brought together experts from many different fields (and 11 partner institutions) in order get a complete picture of the environment impact. Within SHEBA the fate of pollutants into water and air emitted by sea going ships and~~

ships at berth was assessed, both for the present-day situation as well as for several developed scenarios of shipping in the intermediate future. The task was building mainly on a suite of consecutive models following pollutants on the path from emission via dispersion to impacts in target regions.

As part of the SHEBA project, the transport, chemical transformation and fate of atmospheric pollutants in the Baltic Sea region was simulated with three different regional CTM systems (CMAQ, EMEP/MSC-W and SILAM) to investigate the effect of ship emissions on the regional air quality in the Baltic Sea region. The [EMEP/MSC-W model \(MET Norway\)](#) is also included with the same model configuration in the Baltic Sea region Interreg project EnviSuM (Environmental Impact of Low Emission Shipping: Measurements and Modelling Strategies). The main focus of the EnviSuM project is on sulphur emissions from shipping. EnviSuM investigates the effects of the implementation of the stricter SECA from 2015 onwards, combining measurements and modelling. Prank et al. (2016) evaluated the skill of air quality models including SILAM, EMEP, and CMAQ to reproduce the particulate matter concentration and composition on European scale. The chosen CTM systems are well established in Europe and have been extensively tested in several multi-model assessment studies (Solazzo et al., 2012a, 2012b, 2013, 2017; Vautard et al., 2012; Colette et al., 2011, 2012; Langner et al., 2012; Vivanco et al., 2018). Prank et al. (2016) evaluated the skill of air quality models including SILAM, EMEP, and CMAQ to reproduce the particulate matter concentration and composition on European scale. EMEP/MSC-W (from MET Norway) and SILAM (from FMI) are part of the operational Copernicus Atmosphere Monitoring Service (CAMS;) regional air quality ensemble for Europe. All three models have been used previously in the North Sea and Baltic Sea region for estimating the effect of shipping (Jonson et al., 2015; Antturi et al., 2016; Matthias et al., 2010, 2016). The model setup with CMAQ used in Matthias et al. (2016) has been evaluated for the larger North Sea region (Aulinger et al., 2016).

The EMEP/MSC-W model (at MET Norway) is also included with the same model configuration in the Baltic Sea region Interreg project EnviSuM (Environmental Impact of Low Emission Shipping: Measurements and Modelling Strategies). The main focus of the EnviSuM project is on sulphur emissions from shipping. EnviSuM investigates the effects of the implementation of the stricter SECA from 2015 onwards, combining measurements and modelling. This includes measurements of emissions from ships applying different technologies as for instance low sulphur oil, scrubber technology, and liquefied natural gas (LNG). The improved emission estimates are used in chemical tracer models calculating the effects on air pollution and depositions and subsequent effects on human health and ecosystems.

This study takes a multi-model approach using three CTM systems to assess the uncertainties connected with the atmospheric transport and transformation of air pollutants. The comparison of air concentration of regulatory pollutants between the models is the primary focus of this study. However, the annual average nitrogen deposition fields were also compared due to the relevance of atmospheric nitrogen deposition for the eutrophication of the coastal marine environment. In the western Baltic Sea, with its specific coastal features such as boddens and lagoons, the grid resolution of atmospheric CTMs has been found to strongly impact on the modelled nitrogen deposition (Neumann et al., 2018a). Model results from the three CTMs for [Modelled total air concentrations of NO_x, O₃, SO₂ and PM_{2.5} from the three CTMs](#) are compared to observations from rural and urban background stations in the coastal areas of the Baltic Sea region. Statistical performance analysis of the comparison of modelled against observation data [of total concentrations](#) was carried out for all CTM systems and the performance of the models was

inter-compared based on several statistical indicators. Specifically, we want to evaluate the contribution of shipping emissions to modelled surface concentrations of important air pollutants. The significance of ship contribution to ambient NO₂ observations at coastal monitoring stations is evaluated for the different models. The use of three CTM systems, together with comparison to ground-based observations, provides a comprehensive view on the current air quality situation of the Baltic Sea region and how it is affected by emissions from shipping. The combination of three CTM also provides a more robust estimate of the ship-related contribution to ambient atmospheric concentrations.

2 Description of the CTM systems and setup of the model comparison

The setup of the three CTM systems for this study with respect to drivers for meteorology, boundary conditions and emissions was specific for each model system. The models were set up in a way as they are typically used in air quality studies in European regions. However, the applied CTMs use a much higher spatial and temporal resolution as previous modelling of the air quality in the Baltic Sea region. Shipping activities are considered with a high degree of detail in the simulations; using [AIS-Automatic Identification System \(AIS\)](#) position data and up-to-date load dependent emission factors for all important air pollutants. The dynamic ship emission inventory [STEAM-Ship Traffic Emission Assessment Model \(STEAM; Jalkanen et al., 2009, 2012\)](#) was applied in all CTMs. STEAM takes into account the emission control areas and regulations, emission abatement equipment on-board the ships as well as fuel sulphur content modelling separately for main and auxiliary engines (Johansson et al., 2017; Jalkanen et al., 2012). All three regional air quality models implement state-of-the-art formulations of atmospheric transport, atmospheric chemistry and aerosol formation, updated compared to the model versions used in the previous studies. Partly the same or similar drivers for anthropogenic emissions were used in the CTMs. Ship exhaust emission from the North Sea are handled in the same manner as the Baltic Sea emissions since they affect the western part of the Baltic Sea region. By this procedure it is ensured, that all interactions of shipping emissions with pollutants in the regional background and from land-based emission sources are correctly considered. With all models a reference run for the current air quality situation was performed including all emissions (“base”) and one run without the emissions from shipping (“noship”). The difference between the run with all emissions and the run without shipping emissions is used to determine the contribution of ships to the ambient pollutant concentration. [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 \(Jonson et al., 2018a; Karl et al., 2019\).](#)

2.1 Description of the models

2.1.1 CMAQ model

The CMAQ model v5.0.1 (Byun and Schere, 2006; Appel et al., 2013; 2017) computes the air concentration and deposition fluxes of atmospheric gases and aerosols as a consequence of emission, transport and chemical transformation. The atmospheric chemistry is treated by the modified Carbon Bond V mechanism cb05tucl with updated toluene chemistry and chlorine radical

chemistry (Yarwood et al., 2005; Whitten et al., 2010; Sarwar et al., 2012). The aerosol scheme AERO5 is used for the formation of secondary inorganic aerosol (SIA). The gas phase/aerosol equilibrium partitioning of sulphuric acid (H_2SO_4), nitric acid (HNO_3), hydrochloric acid (HCl) and ammonia (NH_3) is solved with the ISORROPIA v1.7 mechanism (Fountoukis et al., 2007; Nenes et al., 1999). The formation of secondary organic aerosol (SOA) from isoprene, monoterpenes, sesquiterpenes, benzene, toluene, xylene, and alkanes (Carlton et al., 2010; Pye and Pouliot, 2012) is included.

The dry deposition parameterization is presented in Binkowski and Shankar (1995) and Binkowski and Roselle (2003). Wet deposition of gases and particles is computed by the resolved cloud model of CMAQ which estimates how much certain vertical model layers contributed to the precipitation (Foley et al., 2010). Sea salt emissions were calculated inline by the parameterization of Gong (2003) (as described in Kelly et al., 2010). Sea salt surf zone emissions were deactivated because of considerable overestimations in some coastal regions (Neumann et al., 2016). Biogenic emissions (NMVOC from vegetation and soil NO) were calculated off-line with the biogenic Emission Inventory System BEIS v3.4 (Schwede et al., 2005; Vukovich and Pierce, 2002). Emissions of wind-blown dust were not considered.

The Meteorology-Chemistry Interface Processor (MCIP; Otte and Pleim, 2010) processes meteorological model output into the input format required for CMAQ. The vertical dimension of the model extends up to 100 hPa in a sigma hybrid pressure coordinate system with 30 layers. Twenty of these layers are below approximately 2 km; the lowest layer extends to ca. 36 m above ground. A spin-up period of one month (December 2011) was used for the initialization of the model runs, sufficiently long to prevent that initial conditions have an effect on the simulated atmospheric concentrations of the investigated period (year 2012).

2.1.2 SILAM model

The SILAM (System for Integrated modelLling of Atmospheric coMposition) model v5.5 (Sofiev et al., 2015; <http://silam.fmi.fi/>) was used as second CTM in this study. The gas phase chemistry was simulated with the Carbon Bond (CB) mechanism CBM-IV, with reaction rates updated according to the recommendations of the International Union of Pure and Applied Chemistry (IUPAC, <http://iupac.pole-ether.fr>) and the NASA Jet Propulsion Laboratory (JPL; <http://jpldataeval.jpl.nasa.gov>) and the terpenes oxidation added from CB05 reaction list (Yarwood et al., 2005). The sulphur chemistry and secondary inorganic aerosol formation is computed with an updated version of the DMAT scheme (Sofiev, 2000) and secondary organic aerosol formation with the Volatility Basis Set (VBS, Donahue et al., 2011), the volatility distribution of anthropogenic organic carbon taken from Shrivastava et al. (2011). Organic aerosol in SILAM was evaluated in a recent study by Prank et al. (2018).

The dry deposition scheme is described in Kouznetsov and Sofiev (2012) and the wet deposition in Kouznetsov and Sofiev (2013). Natural emissions included in the simulations are sea-salt emissions as in Sofiev et al. (2011), biogenic NMVOC emissions as in Poupkou et al. (2010); wild-land fire emissions as in Soares et al. (2015) and wind-blown desert dust.

SILAM includes a meteorological pre-processor for diagnosing the basic features of the boundary layer and the free troposphere from the meteorological fields provided by various meteorological models (Sofiev et al., 2010). In total 10 vertical layers, extending up to 2000 m above the surface, are included. The lowest layer extends to 20 m above the surface. No spin-up period was applied.

2.1.3 EMEP/MSC-W model

The third CTM used in this study is the EMEP/MSC-W chemical transport model, version rv4.8. This model, available as open source (<https://github.com/metno/emep-ctm>), has been described in detail in Simpson et al. (2012), with various updates, see Simpson et al. (2016) and references within. Chemistry scheme of the gas-phase in the model is EmChem09, having 72 chemical compounds including 10 “surrogate” VOCs, out of which isoprene represents BVOCs, and 137 reactions. This scheme is an update of previous chemical schemes (e.g. Simpson, 1992; Andersson-Sköld and Simpson, 1999). The EMEP scheme involves relatively more details on peroxy radical (RO₂) chemistry than e.g. Carbon Bond (CB) schemes. SOA is calculated using a VBS scheme, which tracks the semi volatile products of VOC oxidation, and dynamically partitions these between the gas and aerosol phases (e.g. Robinson et al., 2007). A number of schemes were tested in Bergström et al. (2012), but here the standard “NPAS” scheme as described in Simpson et al. (2012) is used.

For Europe the model is regularly evaluated against measurements in the EMEP annual reports, see www.emep.int. In addition the EMEP model has been included in model inter-comparisons and model validations in a number of peer reviewed publications (Jonson et al., 2006, 2010, 2018a; Simpson et al., 2006a, 2006b; Colette et al., 2011, 2012; Angelbratt et al., 2011; Dore et al., 2015; Stjern et al., 2016). Biogenic emissions (NMVOC, soil NO), emissions of dimethyl sulphide (DMS) from oceans, sea-salt, dust, road dust, emissions from aviation on cruising altitude, lightning, volcanic emissions and emissions from forest fires are included as separate databases or calculated within the model (EMEP, 2015).

EMEP is driven by the meteorological data of the European Centre for Medium-Range Weather Forecasts (ECMWF) based on the Cy40r1 version. An important addition to the forecast ensemble in cycle Cy40r1 has been the introduction of ocean-atmosphere coupling from day 0, instead of from day 10 as in the previous cycles. Vertically, the meteorological fields from ECMWF are interpolated onto 20 EMEP sigma levels, between the surface and 100 hPa. Initial and boundary concentrations are based on long-term observations and some model data. No spin-up period was applied.

2.2 Setup of the CTM systems

The CTMs were offline coupled with different meteorological models (COSMO-CLM, ECMWF-IFS, and WRF). CMAQ and SILAM were operated with high horizontal resolution (4 km) on the inner nest representing the Baltic Sea region starting from simulations of a coarser European domain. EMEP MSC-W was operated on 0.1 degree resolution for the whole of Europe. Ship emissions from the STEAM model (Jalkanen et al., 2009; Jalkanen et al., 2012; Johansson et al., 2013) ~~,-which use ship position data of the HELCOM AIS network,~~ were gridded to the respective model's grid resolution. Land-based emissions were from SMOKE-EU (Bieser et al., 2011a) or ECLIPSE (Amann et al., 2012; Amann et al., 2013) databases; annual totals were comparable.

2.2.1 Model domains and nesting

The spatial extent for the intercomparison study covers the Baltic Sea region, spanning from latitude 53.50°N (south) to 66.00°N (north) and longitude 9.00°E (west) to 31.00°E (east). Parts of the Kattegat and a small part of the Norwegian Sea is

covered by the extent, but not considered in the comparison. The extent of the geographic domain is displayed in Fig. 1. Nested simulations were done with CMAQ and SILAM models, using the output of the finer resolved inner nest whereas the simulation with the EMEP/MSC-W model covered the European domain. The SILAM model was operated on rotated grids centred on the respective domain. The horizontal grid resolution of the output was 4 km for CMAQ, 0.04 degrees (~4 km) for SILAM and 0.1 degrees (~11 km) for EMEP/MSC-W. Note that the grid distance in x-direction becomes smaller with increasing latitude (for instance, 0.1° in longitude corresponds to 6.2 km at 56°N). ~~Different drivers were used for meteorological simulations coupled offline to the CTM simulations.~~ Anthropogenic emissions from the continent and the shipping emissions in the North and Baltic seas were identical (CMAQ and SILAM) or similar in spatial distribution and magnitude (EMEP/MSC-W). The EMEP/MSC-W model used monthly averaged gridded ship emissions, while the other two models used hourly emissions. Daily or hourly emissions reflect ship traffic pattern changes due to meteorological conditions or due to sea ice. Using a coarser time resolution for shipping thus mainly neglects the influence of weather and ice on ship operations (Jonson et al., 2015). Table 1 gives an overview of the model setups of the three CTM systems for use in the intercomparison study.

Nested simulations with CMAQ were performed with a coarse outer domain for entire Europe with grid cell size of $64 \times 64 \text{ km}^2$, an intermediate domain with $16 \times 16 \text{ km}^2$ for Northern Europe and an inner domain with a horizontal resolution of $4 \times 4 \text{ km}^2$ for the entire Baltic Sea. Model results for the intercomparison were taken from the inner domain for the coastal regions and from the intermediate domain for parts of Sweden, Finland and the Baltic states. For details on the high-resolution output from CMAQ and an evaluation of the model setup with a limited number of regional background stations, it is referred to [Karl et al. \(2018\)](#) [Karl et al. \(2019\)](#).

For the SILAM model, the grid cell size was roughly $70 \times 70 \text{ km}^2$ for the outer domain, roughly $18 \times 18 \text{ km}^2$ for the central domain, and roughly $4 \times 4 \text{ km}^2$ for the inner domain. The simulation time steps were 20 min, 10 min, and 4 min, respectively. Model results for the intercomparison were mostly from the inner domain, with parts of Finland and eastern Europe from the central domain. SILAM took part in AQMEII 1 and 3 intercomparisons showing comparable performance with other European state-of-the-art air quality models (Solazzo et al., 2012a, 2012b, 2013, 2017; Vivanco et al., 2018; Marécal et al., 2015). The EMEP/MSC-W model was run with a 0.1×0.1 degrees resolution for whole Europe. A comprehensive description, including model evaluations, of the model results with the 0.1×0.1 degrees application of the EMEP model for 2013 can be found in Tsyro et al. (2015).

2.2.2 Meteorology

The SILAM model is run with meteorological input from a simulation with the Weather Research and Forecast (WRF) model v3.7.1 using original resolutions of 4.0 km, 16.0 km, and 64.0 km, for inner, central and outer domains, respectively. [WRF was driven with large scale meteorological forcing data taken from the ERA-Interim reanalysis \(Dee et al., 2011\).](#) In general, linear interpolation was applied for the simulation, but conservation of mass was used where applicable. [The high-resolution inner domain extended up to 2000 m height and was therefore less influenced by upper tropospheric dynamics of WRF. Kryza et al. \(2017\), using WRF in a similar configuration, evaluated the WRF meteorological fields against station observations in Poland. The 2 m air temperature \(T2\) was underestimated in winter \(bias smaller than -0.6 K\) while temperature](#)

in the warm season was overestimated (bias up to +1.0 K). The largest errors for the 10 m wind speed (WS10) occurred in late summer and autumn and the largest errors for wind direction in spring and summer. The error of wind direction was very small in winter. Spatial distribution of meteorological variables obtained from WRF were in close agreement with the station measurements, but the model performance was found to be worse for the seashore and mountain areas than for other inland areas (Kryza et al., 2017).

High-resolution meteorological fields for CMAQ were obtained from the COSMO-CLM (Rockel et al., 2008) model v5.0. ~~More details on the meteorological forcing data and the evaluation of precipitation can be found in Karl et al. (2018).~~ The meteorological fields were converted to the extension, resolution and projection of the CMAQ nested grids, using an in-house modified version of MCIP. ~~More details on the meteorological forcing data and the evaluation of precipitation can be found in Karl et al. (2019).~~ Here we include an evaluation of T2 and WS10 in the southern part of the Baltic Sea region. Temperature was compared against gridded observational dataset E-OBS v.16 (Cornes et al., 2018). Wind speed was compared against observational data from MiKlip DecReg of the German Weather Service (DWD). Monthly mean T2 in Denmark and southern Sweden was underestimated in winter (bias smaller than -1.4 K) and overestimated in summer. The warm bias in summer was higher in Sweden (+1.4 K) than in Denmark (+0.4 K). The spatial correlation of T2 in the southern Baltic Sea region based on 3-daily averages was remarkably good. Monthly mean WS10 was slightly overestimated in most parts of the region. The largest errors of wind speed occurred in Denmark and northern Poland during May and June.

EMEP/MSC-W was driven by meteorological data from the Integrated Forecasting System (IFS) of the ECMWF, version IFS38r2, with t1279 resolution (about 0.16 degrees resolution) interpolated to 0.1×0.1 degrees. ~~The ECMWF forecasting system of weather parameters is regularly validated by comparing against European synoptic observation data available on the Global Telecommunication System (GTS).~~ The evaluation of the weather forecast from cycle Cy40r1 is summarized as follows (Haiden et al, 2014). The frequency of light precipitation is overestimated, with a bias of $1.2\text{--}1.4 \text{ mm d}^{-1}$ (for precipitation amounts $> 1 \text{ mm d}^{-1}$). T2 has a negative night-time temperature bias over Europe in winter and early spring. For total cloudiness, bias and standard deviation are small in 2012. For WS10, the standard deviation is low and the night-time bias is very small.

The use of different meteorological datasets introduces additional variability which is on one hand wanted to achieve a wider range of possible results for estimating the effect of shipping on air quality but on the other hand complicates the interpretation of differences between the models.

2.2.3 Boundary conditions

The initial conditions (ICONS) for the simulation and the lateral boundary conditions (BCONs) for the outer European domain are taken from FMI APTA global reanalysis (Sofiev et al., 2018a)). The global boundary conditions results have been interpolated in time and space to provide hourly boundary conditions for the respective outer domains of the CMAQ and SILAM simulations. The setup for initial and boundary concentrations for EMEP/MSC-W is described in Simpson et al. (2012). ICONs and BCONs are based on long-term observations. For ozone, 3-D fields for the whole domain are specified from climatological ozone sonde datasets, modified monthly against clean-air surface observation. For most other chemical compounds they are

defined by simple functions based on measurements and/or model calculations, prescribing concentrations in terms of latitude and time-of-year, or time-of-day.

2.2.4 Anthropogenic land-based emissions

Anthropogenic land-based emissions in hourly resolution obtained from the SMOKE-EU (Bieser et al., 2011a) emission inventory were provided for CMAQ and SILAM. These emissions are based on officially reported EMEP emissions which are then distributed in time and space using appropriate surrogates like population density maps, street maps or land use maps. Point sources from the European point source emission register are considered. Vertical distribution of point source emissions is based on real-world stack information and calculated within SMOKE-EU (Bieser et al., 2011b). Dynamic emissions from agricultural activity and animal husbandry depending on meteorological variability are considered (Backes et al., 2016). EMEP/MSC-W model uses anthropogenic emissions from the ECLIPSE gridded emission inventory (<http://www.iiasa.ac.at/web/home/research/researchPrograms/air/ECLIPSEv5.html>). These emissions differ slightly from the reported national total EMEP emissions for 2012, see Wankmüller and Mareckova (2014). For the countries bordering the Baltic Sea (excluding Russia) the national total sulphur emissions from ECLIPSE are about 6 % higher and the NO₂ emissions about 10 % lower than the corresponding EMEP emissions.

2.2.5 Shipping emissions

~~Shipping emissions for the Baltic Sea and North Sea areas were obtained from STEAM (Jalkanen et al., 2009, 2012; Johansson et al., 2013, 2017). The shipping emissions are used together with the land-based emissions described in Sect. 2.2.4 in all three CTM systems. STEAM uses ship position data of individual ships collected from the AIS base stations network. The AIS data is combined with technical information of the specific vessels and engine types to derive emissions for each individual ship. Shipping emissions are calculated dynamically to consider the emission control areas and regulations, emission abatement equipment on-board the ships as well as fuel sulphur content modelling, separately for main and auxiliary engines (Johansson et al., 2017; Jalkanen et al., 2012).~~

The STEAM inventory for the Baltic Sea shipping emissions used in the SHEBA project, consist of hourly updated $2 \times 2 \text{ km}^2$ gridded data for NO_x, SO_x, carbon monoxide (CO), and particulate matter, which is further divided into elemental carbon, organic carbon, sulphate (SO₄²⁻) and mineral ash. For the North Sea and other European seas the STEAM data for 2011 were used. Ship emission were used with hourly time resolution in CMAQ and SILAM whereas they were used with monthly resolution in EMEP/MSC-W.

The use of monthly aggregated ship emissions in EMEP/MSC-W is justified by the fact that the same set of ship emissions from FMI is applied for different meteorological years in the routine application of EMEP modelling and that ship emissions from other seas were only available for 2011. Previous tests with daily and monthly aggregated ship emissions showed that the differences in results are very small. The use of North Sea ship emissions from 2011 on hourly basis in CMAQ and SILAM causes some inconsistency because meteorological data of 2012 is used in the CTM simulations. Because we are

mainly interested in the seasonal variability of pollutant concentrations based on daily averages, the outcome of this study will be less affected by the inconsistency between the timing of ship emissions and the meteorological conditions.

STEAM emission data was provided for two vertical layers (below 36 m, from ~~36–1000~~36–100 m). ~~The vertical distribution of STEAM emissions was applied differently in the CTMs (see Table 2). In the CMAQ model~~In CMAQ and SILAM, emissions below 36 m were attributed to the ~~lowest vertical model layer~~vertical model layers below 42 m height, while emissions above 36 m were attributed to the ~~second lowest layer. In the SILAM simulation, the emissions were assumed to be evenly distributed within the emission layers between ground and 1000, and were attributed to the model layers accordingly~~model layers between 42 m and 84 m. In EMEP/MSC-W all ship emissions were attributed to the lowest vertical model layer, which typically has a height of 92 m.

10 2.3 Statistical analysis

2.3.1 Evaluation method for the total air pollutant concentrations

Model results for total surface concentrations of NO₂, O₃, SO₂, and PM_{2.5} from the three CTMs are evaluated against available measurements of the air quality monitoring network from the AirBase version 8 database (Simoens, 2014). AirBase is the air quality information system maintained by the European Environmental Agency (EEA) through the European topic centre on
15 Air Pollution and Climate Change Mitigation.

Table S1 gives a list of all rural and regional background monitoring stations. Concentrations of NO₂ are monitored at 17 stations, O₃ at 35, SO₂ at 11, and PM_{2.5} at 8 rural/regional background stations. Table S2 gives all urban and suburban background monitoring stations included in the statistical evaluation of the models. Concentrations of NO₂, O₃, SO₂, and PM_{2.5} are monitored at 52, 46, 37, and 10 stations of the urban background, respectively. Fig. 2a shows locations of stations
20 with NO₂ and with O₃ measurements. Fig. 2b shows locations of the stations with SO₂ and with PM_{2.5} measurements.

The model output of surface concentration fields of each CTM is used with its original horizontal resolution to calculate daily mean concentrations. The modelled concentrations are extracted from the respective model grid cell where the selected monitoring stations are located. The evaluation was done for the entire year 2012 based on daily means. The model output for PM_{2.5} was taken from the modelled PM_{2.5} containing aerosol water at 50 % relative humidity.

25 The performance of each model is quantified in terms of mean values (μ_{Mod} and μ_{Obs}), normalized mean bias (NMB), Spearman's correlation coefficient (R), root mean square error of the modelled values (RMSE) and fraction of model values within factor 2 of the observations (FAC2). Definitions of NMB, R, RMSE and FAC2 are given in Appendix A. The model performance analysis is discussed separately for rural background stations and urban background stations. In order to better highlight model differences in terms of urban areas and station types (i.e. rural, sub-urban, urban background sites), groups
30 of stations (rural versus urban) are generated in which statistical performance indicators are averaged. In the rural group, rural background and regional background stations are included, while in the urban group, urban background and sub-urban background stations are included. Monitoring stations classified as traffic stations and industrial stations were not included in the comparison, since the regional CTM systems applied here do not handle the local scale dispersion near emission sources.

In the context of this evaluation of predicted air pollutant concentrations, we consider a correlation coefficient of more than 0.5 to indicate a correlation between modelled and observed time series, while values of 0.7 and above are considered as a good correlation. Hanna and Chang (2012) define certain acceptance criteria for model performance based on their experience in conducting a large number of model evaluation exercises. For rural stations FAC2 values > 0.5 and for urban stations FAC2 values > 0.3 indicate acceptable performance. We adopt these bounds in the present study to characterize the predictive strength of the models with respect to the pollutant concentrations.

~~Further, we~~ We compare the performance between models with the help of a graphical comparison in form of boxplots. Boxplots of the correlation coefficient, NMB and RMSE including either all rural or all urban monitoring stations were prepared. The boxplots show the median as line dividing the box in two parts, the upper and lower quartiles as end of the box, the minimum and maximum values of the data and outliers.

In addition to the model performance for temporal correlation we evaluated the spatial correlation of the total air pollutant concentrations with the observations of the AirBase network for the three CTMs.

2.3.2 Significance of the ship contribution

The method described in Aulinger et al. (2016) was used to assess the significance of ship influence on ambient NO_2 at the monitoring stations. The ship influence at a station was positively confirmed in the tests if: (1) the concentrations increased and (2) the temporal correlation improved, when shipping emissions are included in the CTM simulation.

By means of a paired t-test it was first tested whether the modelled NO_2 concentrations at the monitoring stations with available NO_2 observations (Table S1) significantly increased if shipping emissions were considered. This test estimated whether the mean concentration difference between the “noship” run and the “base” run (“~~noship~~” - “base”) is significantly equal to or greater than zero, indicated by the probability p_{bias} . If the value of p_{bias} ~~is larger~~ was greater than the level of significance of 0.05, than this hypothesis was confirmed. ~~Otherwise, which means that no difference between the “base” run and the “noship” run could be statistically proven. In case p_{bias} was less than 0.05,~~ it was decided that the model run without shipping emissions led to lower concentrations, confirming the ship influence.

The significance of the improvement in the correlation between simulations and observations was tested by calculating the Fisher z transformation of the two correlation coefficients for the two model runs (“noship” and “base”) and testing the hypothesis “greater than”. Correlation coefficients were calculated with Spearman's method (Myers and Sirois, 2006) for consistency with the statistical evaluation. The probability p_{corr} for the hypothesis that the correlation between the base run and observations is greater than the correlation between the “noship” run and observations was calculated. We accepted this hypothesis if the probability was higher than 0.9. Therefore, in the following, a station i with $p_{\text{corr},i} > 0.9$ for a specific CTM simulation is termed ship-influenced.

3 Results

3.1 ~~Comparison to observations~~ Statistical evaluation of air pollutant concentrations

3.1.1 Rural versus urban sites

A statistical performance analysis for each of the three CTMs was undertaken using the available observation data from AirBase for 2012 based on daily mean total concentrations. The ~~results of the statistical performance analysis~~ evaluation results for the temporal variation of air pollutants are summarized in Table S3 for daily mean NO₂, in Table S4 for daily mean O₃, in Table S5 for daily mean SO₂ and in Table ~~3-S6~~ for daily mean PM_{2.5}. In the following, the performance of the models to simulate air pollutant concentrations is compared and discussed separately for the group of rural stations and for the group of urban stations in order to highlight differences in the predictive capability of the models for rural versus urban sites. ~~Moreover, the significance of the ship contribution to the monitored daily mean has been tested for the three CTMs. The results of the significance test are summarized in Table S6.~~

3.1.1 ~~Statistical evaluation for NO₂ at rural and urban sites~~

~~Emissions of from traffic and various combustion sources are mainly in the form of nitrogen oxide (NO) which oxidises to within a few minutes through the titration reaction with O₃. NO₂ plays a major role in atmospheric reaction cycles that produce ground-level ozone. also contributes to the formation of secondary particulate matter.~~ The atmospheric lifetime of NO₂ is relatively short; a few hours in summer and up to one day in winter (Schaub et al., 2007), hence differences between rural and urban sites are expected due to the higher emission density in urban or industrial areas.

~~Fig. S1 compares the times series of daily mean concentrations from the three CTMs at two rural background stations and at two urban background stations. The seasonal variation at the two rural sites DESH008 (Bornhöved, Germany) and DK0054A (Keldsnor/9054, Denmark) with peak concentrations in winter are captured by the models (Fig. S1a,b). CMAQ shows frequent peaks at the DK0054A during summer. This feature is also seen for other coastal rural sites and could indicate that, due to the high spatial resolution of the model, ship exhaust plumes are resolved but not adequately dispersed. Since the models do not specifically treat the plume dispersion of individual ships, the spreading of the plume might not be sufficiently large or the plume rise of ship exhaust is not properly considered with the applied vertical profile of ship emissions. The other two models also tend to give higher than observed at DK0054A in summer. The correlation between predicted and observed daily mean for this site is low for all models (R < 0.3). The seasonal variation at the two urban sites DK0053A (Aalborg/8158, Denmark) and LVORKE2 (Riga-Kengarags-2, Latvia) are reproduced by the models, although CMAQ and SILAM underestimate daily mean in winter at LVORKE2 (Fig. S1c,d).~~

~~The rural station average of observed annual mean is 6.9 (range: 1.1–15.8). The models reproduced the annual means at rural sites very well; modelled annual mean is 6.5, 8.1 and 6.0 for CMAQ, SILAM and EMEP/MSC-W, respectively. The overall correlation of for rural stations is good for all models, with R of 0.72, 0.64 and 0.65 for CMAQ, SILAM and EMEP/MSC-W, respectively, as shown in the boxplots in Fig. 3a. At most rural stations the NMB is between -0.5 and 0.5 for all models. An~~

exception is SE066A (Norr Malma, Sweden), where is largely overestimated by the models (outlier in the boxplot). This is also the only rural station, for which FAC2 of all models is below 0.5. For SE066A, the influence of traffic emissions from the nearby motorway might be too high in the models, as the spatial resolution is still not fine enough to resolve the dispersion near local sources. The average RMSE at rural sites is 4.7, 5.9 and 3.9 for CMAQ, SILAM and EMEP/MSC-W, respectively.

5 The urban station average of observed annual mean NO₂ is 16.7 (range: 6.7–34.1), more than twice the concentration average at rural sites. The three CTMs underestimate the annual means at ~~urban sites (range of model averages: 9.9–10.9)~~ the urban sites. The overall correlation of NO₂ for rural stations is good for all models. The overall correlation of NO₂ for urban sites is lower than at the rural sites, ~~with R between 0.5 and 0.6 for the models, as shown in the boxplots in (Fig. 3b)~~. At most urban stations, models underestimate the observed daily mean NO₂ by ca. 40 % (NMB ranges: ~~–0.9–0.1, –0.8–0.2 and –0.8–0.4~~ for CMAQ, SILAM and EMEP/MSC-W, respectively) (Table S3). ~~For some urban stations the FAC2 is below 0.3, but for the majority of the stations all models fulfil the acceptance criteria for urban sites. The average RMSE at urban sites is 9.9, 10.1 and 9.6 for CMAQ, SILAM and EMEP/MSC-W, respectively.~~

The general underestimation of NO₂ at urban sites has been evident in other multi-model air quality studies in Europe (e.g. Giordano et al., 2015). The finer horizontal resolution of CMAQ and SILAM (4 km) compared to EMEP/MSC-W (11 km)
15 does not result in a significant improvement of the urban bias and urban temporal correlation. This result was expected based on the study by Schaap et al. (2015), who found no further improvement of the urban signal, i.e. the concentration difference between high emission areas and their surroundings, when increasing the resolution from 14 km to 7 km in the same model. Moreover, increasing the spatial resolution in the model does not help to improve significantly the performance in time because the temporal variability of pollutants is largely controlled affected by the meteorological conditions and pollution levels upwind
20 (Schaap et al., 2015).

3.1.2 Statistical evaluation for O₃ at rural and urban sites

Tropospheric ozone is largely controlled by the atmospheric transport from regions outside the study area, by stratosphere-troposphere exchange and by the photochemical production through the oxidation of VOCs and carbon monoxide (CO) in the presence of NO_x and sunlight. ~~Near the surface, the depletion via reaction with (titration reaction) in environments with high~~
25 ~~levels and dry deposition are important sinks of ozone.~~

~~The seasonal variation at the two rural sites DESH008 and DK0054A (same stations as for) is captured by the models, but high concentrations in July and August at DK0054A are underestimated by CMAQ and SILAM (Fig. S2a,b). The underestimation of observed ozone in summer by the CMAQ simulation is also found for other coastal stations (Karl et al., 2018) and indicates too low photochemical reactivity. The seasonal variation at the two urban sites DK0053A and LVORKE2 is captured by the~~
30 ~~models (Fig. S2c,d). The good performance of the models for at the urban sites is partly attributed to the high spatial resolution, as emissions are injected into a smaller grid box volume and consequently less diluted initially. Thus the models are capable of representing the titration effect on the day-to-day variation of ozone.~~

~~The rural station average of observed annual mean is 56 (range: 44–71). The models show a good correlation with observed at the rural stations is, with R of 0.74, 0.76 and 0.77 for CMAQ, SILAM and EMEP/MSC-W (Fig. 4a). Modelled daily mean is~~

in agreement with observed daily mean within a narrow NMB range for rural sites. CMAQ and SILAM have no bias (average NMB = 0.0), whereas EMEP/MSC-W slightly overestimates the measurements (average NMB = 0.17). The ozone bias might be linked to boundary conditions (Giordano et al., 2015): EMEP/MSC-W uses ozone boundary conditions from long-term observations, whereas CMAQ and SILAM models use boundary conditions from the FMI APTA global reanalysis.

5 The higher density of NO_x emissions in urban areas is expected to lead to a larger titration effect of NO on ozone, which results in lower average O₃ at the urban sites compared to rural sites. The ~~urban station average of observed annual mean models slightly overestimate the O₃ is 48 (range: 38–59) measurements at urban sites, with CMAQ having the smallest bias.~~ CMAQ and SILAM predict similar annual mean concentrations as observed for both rural and urban sites, whereas ~~EMEP/MSC-W EMEP/MSC-W~~ predicts higher annual mean ozone ~~As for the rural sites, the models show a good correlation with observed at the urban stations, with R between 0.73 and 0.77 (Fig. 4b).~~ The ozone bias might be linked to boundary conditions (Giordano et al., 2015): the EMEP model uses ozone boundary conditions from long-term observations, whereas CMAQ and SILAM models use boundary conditions from the FMI APTA global reanalysis.

The models slightly overestimate the O₃ measurements at urban sites, with CMAQ having the smallest bias (average NMB = 0.08). ~~FAC2 of the models is larger than 0.75 for all rural sites and larger than 0.57 for all urban sites.~~ The average RMSE values for the rural sites and the urban sites, respectively, are similar for the three models (Fig. 4), indicating comparable model performance for the CTMs with respect to daily mean O₃ concentrations.

3.1.3 Statistical evaluation for SO₂ at rural and urban sites

Another major air pollutant is SO₂. It is primarily emitted from anthropogenic emission sources such as coal power plants, residential heating, waste incineration and shipping activities. SO₂ acts as a precursor to sulphates, which are one of the main components of particulate matter in the atmosphere. The atmospheric lifetime of SO₂ is on the order of a few days (Lee et al., 2011). SO₂ can still be considered to be relatively short-lived and thus less influenced by transport from regions outside the study area. Most emission sources of SO₂ are located in urban areas. In the case of power plants, the emissions of SO₂ are however injected at elevated height and therefore do not directly impact the surface concentrations in the urban area. ~~On the other hand, residential heating emissions of strongly influence surface concentrations in urban areas during winter. In coastal harbour cities, from shipping is a relevant contributor (Viana et al., 2014).~~

The seasonal variation at the two rural sites SE0001A (Rådhuset, Sweden) and FI00208 (Luuki, Finland; rural) is rather well reproduced by the models (Fig. S3a,b). CMAQ and SILAM overestimate the observed concentrations in winter. EMEP/MSC-W underestimates observed concentrations at FI00208 and also at other rural stations. The seasonal variation at the two urban sites DESH023 (Lübeck-St. Jürgen, Germany) and LT00044 (Kaunas-Noreikiskes, Lithuania) is captured by the models (Fig. S3c,d). CMAQ and SILAM show too high concentrations in winter and EMEP/MSC-W underestimates observed during summer.

The rural The urban station average of observed annual mean SO₂ is 1.3 (range: 0.7–2.5). The annual means of the models at rural sites is in the range of 0.8 to 2.2. With only 10 stations, the rural station group for is rather small, limiting the conclusions that can be drawn from the statistical analysis. The model data and the observations for three times higher than at the rural stations are correlated, with R of 0.67, 0.65 and 0.55 for CMAQ, SILAM and EMEP/MSC-W (Fig. 5a). At rural and urban

sites, the modelled daily mean SO₂ from CMAQ and SILAM has a positive bias, whereas modelled daily mean SO₂ from EMEP/MSC-W has a slight negative bias (Fig. 5). For EMEP/MSC-W some urban stations have a FAC2 of the models is larger than 0.5 for most rural sites. For CMAQ, only one station (EE0016A, Saarejärve, Estonia) does not fulfil the acceptance criteria below 0.3, due to frequent overestimation/underestimation of observed SO₂. The average RMSE at rural sites is 1.5, 2.4 and 1.2 for CMAQ, SILAM and EMEP/MSC-W, respectively.

At urban stations, the temporal correlation between model data and observed SO₂ shows a mixed performance of the models, with good correlation at some stations and poor correlation at others (Fig. 5b). The urban station average of observed annual mean SO₂ is 3.9 (range: 1.0–8.0), three times higher than at the rural stations. CMAQ and SILAM predict similar annual mean concentrations, whereas EMEP/MSC-W predicts 36% lower concentrations for the urban sites. For CMAQ and SILAM, FAC2 is larger than 0.3 for all urban sites. For EMEP/MSC-W some stations have a FAC2 below 0.3, due to underestimation of observed. The average RMSE at urban sites is 4.2 sites are correlated, but with only 10 stations, 4.3 and 3.3 for CMAQ, SILAM and EMEP/MSC-W, respectively. The rural station group for SO₂ is rather small, limiting the conclusions that can be drawn from the statistical analysis. 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.

3.1.4 Statistical evaluation for PM_{2.5} at rural and urban sites

Ambient PM_{2.5} is a wide-spread pollutant, which is directly emitted by biomass and fossil fuel combustion in domestic and industrial activities, and it is also formed from gaseous precursors such as NO_x, SO₂, NH₃ and NMVOC in the atmosphere. The atmospheric lifetime of PM_{2.5} is on the order of days or weeks and thus PM_{2.5} can be subject to long-range transport.

The seasonal variation at the two rural sites SE0001A and FI00208 (same stations as for) is well reproduced by the models (Fig. S4a,b). CMAQ and EMEP/MSC-W underestimate the observed concentrations at SE0001A in summer. The seasonal variation at the two urban sites DESH023 and LT00044 is nicely reproduced by the models (Fig. S4c,d). CMAQ underestimates the observed concentrations at both urban stations in summer. This is also found for other stations of the regional background and can partly be attributed to the underestimation of secondary organic aerosols and to the missing emissions of wind-blown soil dust particles in the CMAQ simulation (Karl et al., 2018).

The rural station average of observed annual mean is 7.0 (range: 3.3–12.6). The modelled annual mean is 5.5, 6.7 and 3.8 for CMAQ, SILAM and EMEP/MSC-W, respectively. For PM_{2.5} smaller differences between rural and urban stations are expected than for NO₂ and SO₂ because PM_{2.5} has a large secondary component, which is generally more homogeneously distributed over rural and urban areas. The urban station average of observed annual mean SILAM is able to reproduce annual mean PM_{2.5} is 10.5 (range: 4.8–13.2) within a similar range as the rural sites. SILAM predicts a similar annual mean concentrations for urban stations, whereas CMAQ and EMEP/MSC-W give lower annual mean values.

For rural stations than observed. For urban stations, the temporal correlation of daily mean PM_{2.5} is good for all models, whereas for rural stations the temporal correlation is slightly better for EMEP/MSC-W than for CMAQ and SILAM (Fig. 6a). For urban stations the temporal correlation of is good, with average R between 0.68 and 0.71 (Fig. 6b). At both rural and urban

sites, the modelled daily mean PM_{2.5} from CMAQ and EMEP/MSC-W has a slightly negative bias, whereas modelled daily mean PM_{2.5} from SILAM has no bias (Table 3).

The average RMSE at the rural sites is between 4.9 and 5.4. It should be noted that RMSE station values for SILAM are within a smaller range (between lower and upper quartile) than the other two models. The performance for urban stations in terms of RMSE is similar for the models, with average RMSE between 6.4 and 7.2

3.1.2 Spatial correlation

The spatial correlation between modelled and observed annual mean total pollutant concentrations for the three CTMs is presented in Fig. 7. Because NO_x is mainly emitted near the ground, the spatial distribution of NO₂ is expected to be highly correlated with emissions. The improvement of spatial resolution of emissions should lead to improved spatial correlation between modelled and observed concentrations. In contrast with this expectation, EMEP/MSC-W shows the best correlation and the lowest bias. Observed annual mean NO₂ at urban stations is strongly underestimated by SILAM and in CMAQ. The positive bias indicates that observed NO₂ at rural stations tends to be overestimated by these two models. The annual mean O₃ is closely linked to the annual mean NO₂ through the local titration effect. Hence lower than observed O₃ at rural stations for SILAM and CMAQ is related to the higher than observed NO₂. The models are capable of representing the seasonal variation of ozone, with highest average concentrations in spring, followed by summer and lowest concentrations in winter and autumn.

The spatial correlation between modelled and observed SO₂ is weaker than that for NO₂ which is probably due to the fact that most SO₂ sources are emitting into higher vertical layers. CMAQ and SILAM overestimate SO₂ in autumn and winter at many stations by a factor of two or more, which is likely related to the uncertainty of residential heating emissions. EMEP/MSC-W underestimates SO₂ in summer which might be connected to uncertainties of the vertical emission distribution. EMEP-MSC/W shows the best spatial correlation for PM_{2.5} with almost no bias. However, annual average PM_{2.5} is underestimated by 23 % on average. For CMAQ and SILAM the spatial correlation for annual mean PM_{2.5} has a positive bias due to overestimation at the rural stations. At almost all stations, CMAQ underestimates PM_{2.5} in summer. This has been evident also for regional background stations of the EMEP monitoring network and can partly be attributed to the underestimation of secondary organic aerosols in the CMAQ simulation (Karl et al., 2019). SILAM underestimates PM_{2.5} in winter. Since PM_{2.5} in winter is mainly from anthropogenic sources and SILAM uses the same emissions as CMAQ, the discrepancy in winter PM_{2.5} is attributed to problems with simulating stagnant meteorological conditions.

3.1.5 Significance of ship contribution to at monitoring stations

3.2 Evaluation of ship-related concentration contributions

A direct comparison of the modelled ship-related concentration contribution to measurements of the shipping signal (in exceedance of the background air) is hampered by the fact that measured concentration increases due to individual ship plumes do not reflect the entire contribution of shipping at sea. In order to evaluate the modelled ship contributions, a statistical method (Sect. 2.3.2) was applied to decide whether the modelled concentration as well as the correlation with observed daily

mean NO₂ concentration at a specific station increases significantly when ship emissions are included in the CTM simulation. ~~If both the concentration and the correlation increased, the station is termed ship-influenced. The results of the significance test are summarized in Table S7.~~

A significant concentration increase was found at all 69 stations for ~~CMAQ and EMEP/MSC-W but only at 78% of the stations for SILAM~~the three CTMs. However, the significance of the concentration increase only shows that the modelled concentrations at a station are sensitive to ship emissions. The correlation increases significantly (on 0.9 or 0.95 level) at ~~7, 10, and 11~~7, and 8 stations for CMAQ, SILAM and EMEP/MSC-W, respectively (Table ~~S6~~S7).

~~The Four ship-influenced stations identified by CMAQ included two stations were identified by all models: Vilsandi (EE0011R, Estonia), Utö (FI00349, Finland) at the shoreline (EE0011R, Vilsandi, Estonia, Fig., Norr Malma (SE0066A), 20 S5a; FI00349, Utö, Finland, Fig.- km inland at the east coast of Sweden, and Lübeck-St. Jürgen, (DESH023, Germany) close to a port (time series plots in Appendix B), S5b), three stations in small~~ At Norr Malma, NO₂ is largely overestimated by the models in summer when ship emissions are included. We suggest that, due to the high spatial resolution of the model, ship exhaust plumes are resolved, but not adequately dispersed in the models. Since the models do not specifically treat the plume dispersion of individual ships, the spreading of the plume might not be sufficiently large or the plume rise of ship exhaust is not properly considered with the applied vertical profile of ship emissions.

Ship-influenced stations found by any of the models included shoreline stations (Virolahti, FI00351, Finland; Lahemaa, EE0009R, Estonia), stations in harbour cities (~~DESH023, Lübeck-St. Jürgen; EE0022A, Narva, Estonia; SE0022A~~Rostock Warnemünde, DEMV021, Germany; Kiel, DESH033, Germany; Aalborg/8158, DK0053A, Denmark, Södermalm, Sweden) and two stations further inland (FI00431, Palokka 2, Finland; PL0077A, KpZielBory TuchSE0022A, Sweden; Gdansk Pm.a09aN, PL0053A; Poland) and one urban inland station (Pm.63.wDSAa, PL0171A, Poland). ~~Some of these stations (EE0011R, FI00349, FI00431 and PL0077A) were also identified by the other models~~The corresponding time series plots of daily mean NO₂ are shown in Appendix B. Observed daily mean NO₂ at the two urban stations in Poland is underestimated by all models, indicating missing local emissions from other sectors. The ship influence at station Rostock Warnemünde(~~DEMV021, Germany~~), located close to a harbour, was significant in both ~~EMEP/MSC-W and SILAM~~EMEP/MSC-W but not in ~~CMAQ~~the other two models (Fig. ~~S5e~~B1i). This could indicate that differences in the meteorological data, in particular wind flow fields, are responsible for the different ship influence. Although, the timing and location of ship exhaust plumes - based on AIS data - should be accurate during the port stays, the emission fluxes at berth are more challenging to estimate, because this involves estimation of electrical power usage during the port stays. ~~Station DK0054A (Keldsnor/9054) (Evaluation of the ship contribution in Rostock using an urban air quality model with high degree of details on ship emissions and other urban emissions showed that shipping significantly impacts on annual averaged NO₂ in the city domain (Ramacher et al., 2019).~~

For all ship-influenced stations, time series plots of daily mean O₃ are compiled in Fig. ~~S1, S5d~~, although close to the shoreline, did not fulfil the correlation criteria, either because the station is not situated downwind of the ship plumes or the models did not properly resolve the ship plumes due to the sub-grid variability of the plume dispersion (see Sect ~~Including the shipping emissions in the model simulations affected ozone concentrations mainly in the summer months. The change of modelled O₃ due to shipping was below 6% on summer average at the ship-influenced stations. At most of the stations,~~

including ship emissions increased the modelled O₃ concentration as a consequence of photochemical ozone production. The seasonal variation of NO₂ at the ship-influenced sites with peak concentrations in winter is in general reproduced by the models. Including ship emissions improved the agreement between modelled and measured NO₂ daily mean concentrations at about 50 % of the stations. For more than 70 % of all stations the observations of total NO₂ concentrations are reproduced by the models within a NMB range from -0.5 to 0.5 in the “base” simulation (Fig. S2).

3.3 Comparison of the spatial distribution of air quality indicators

3.3.1 Spatial distribution of annual mean NO₂

A strong south-north gradient for annual mean NO₂ concentrations is found for the Baltic Sea region in the three “base” simulations, with 4–5 times higher NO₂ concentrations in the south-western part than in the northern part of the region (Fig. 78a). High modelled NO₂ concentrations are predicted in Denmark, northern Germany, and Poland as well as over the Danish Straits and in the urbanized areas of the region. Modelled annual mean NO₂ concentrations in proximity of the main shipping routes several times exceed the concentrations in the regional background. EMEP/MSC-W shows the strongest concentration gradients between urban and rural areas and between ship lanes and surrounding sea. The simulations with the other two models result in a wider spread of the NO_x emissions from the ship routes and the urban centres, indicating stronger horizontal transport by advection and diffusion in CMAQ and SILAM. This finding is counter-intuitive as the NO_x emissions are initially less diluted than in the EMEP/MSC-W simulation because of the smaller volume of the grid boxes and ~~should~~ therefore result in higher NO_x concentrations near the emission sources.

Atmospheric transport by diffusion processes are sub-grid mixing processes, which are not resolved by the given resolution of the applied models. For large grid cells, e.g. 50 × 50 km², the numerical diffusion will usually be much larger than the physical diffusion in the horizontal direction. However, at finer resolution scales, the physical diffusion will gradually become more important than numerical diffusion and becomes greater than numerical diffusion for 5 × 5 km² cell size or below (Karl et al., 2014).

The wider spread of elevated NO₂ concentrations is also indicative for a longer atmospheric lifetime of NO₂ in CMAQ and SILAM compared to the simulation with EMEP/MSC-W. NO₂ is removed relatively quickly in the lower troposphere through the reaction with hydroxyl (OH) radicals to form HNO₃. The rate coefficient for this reaction, $k(\text{NO}_2 + \text{OH})$, is similar in the three models ($(1.1\text{--}1.2) \times 10^{-11} \text{ cm}^3 \text{ s}^{-1}$ at 298 K). Thus, differences in the NO₂ lifetime are mainly due to different abundances of OH radicals in the simulations. High NO₂ concentrations in Belarus and Russia in the SILAM simulation are an artefact from merging with the output of the coarser central model grid (Sect. 2.2.1).

3.3.2 Spatial distribution of annual mean O₃

Modelled annual mean O₃ concentrations over the Baltic Sea are 15–25 % higher than over land, but are reduced along the ship lanes due to the titration effect caused by the ship-emitted NO_x (Fig. 8b7b). Lowest ozone concentrations are seen for St. Petersburg ($< 32 \mu\text{g m}^{-3}$) in the three simulations, although we note that the city is outside of the high-resolution grid in

the case of SILAM. The spatial average of annual mean O₃ is clearly higher for the EMEP/MSC-W simulation, by ~~15–25~~ca. 20 %, compared to the other two simulations (Table 42). The most probable reason for the difference is the application of different sets of boundary conditions for the European model domains, as discussed in Sect. ~~3.1.23.1.1~~. Model simulations for Europe have shown a high sensitivity of ozone changes to the dry deposition to vegetation (Andersson and Engardt, 2010).

5 Thus differences in the deposition schemes may partly explain the different O₃ levels over the continent, e.g. when comparing ozone over Sweden and Finland between CMAQ and SILAM.

3.3.3 Spatial distribution of annual mean SO₂

Clear differences in the spatial distribution of the annual mean SO₂ concentrations are found between CMAQ and SILAM on one hand and EMEP/MSC-W on the other hand (Fig. 78c). The simulation with CMAQ and SILAM show a southeast-northwest

10 gradient with elevated SO₂ over large parts of the southern Baltic Sea region, Poland, Belarus, Russia and the Baltic States with annual mean concentrations in the range of 1.3–3.0 µg m⁻³. Residential heating emissions and power plant emissions for district heating in the urban centres and rural areas strongly contribute to the high SO₂ concentrations in this sub-region. In the EMEP/MSC-W simulation, elevated SO₂ concentrations are present along the main shipping routes, in urban areas and in Poland, whereas the levels of SO₂ outside of these areas are much lower.

15 The concentration gradients between urban and rural areas and between ship lanes and surrounding sea is up to 2.5 µg m⁻³ for EMEP/MSC-W while it is only up to 0.7 µg m⁻³ for CMAQ and SILAM. Factors contributing to the different gradients are differences in the representation of horizontal transport (see Sect. 3.3.1), differences in the meteorological conditions, and differences in the atmospheric lifetime of SO₂. The atmospheric lifetime of SO₂ is determined by its reaction with the OH radical and by its removal via dry deposition. In EMEP/MSC-W, the canopy uptake of SO₂ is strongly controlled by

20 NH₃ levels, and the implemented deposition parameterization accounts for co-deposition effects on the dry deposition of SO₂ (Simpson et al., 2012). Co-deposition effects are not considered in the other two models.

3.3.4 Spatial distribution of annual mean PM_{2.5}

Modelled annual mean PM_{2.5} is higher in the southern part, both over land and sea, than in the northern part of the Baltic Sea region (Fig. 78d). On annual average, PM_{2.5} concentrations are not elevated along the ship routes. The seasonal differences

25 between summer and winter will be discussed below (Sect. 3.5) and will help to understand differences between the models. High PM_{2.5} levels (8 µg m⁻³ and higher) are simulated in the urban areas of major cities like Copenhagen, Oslo, Helsinki, Riga, Tallinn and St. Petersburg. The high PM_{2.5} levels over the continent in the southern part of the Baltic Sea region presumably result from a combination of land-based primary emissions, long-range transported particles and the formation of secondary particulate matter.

30 3.3.5 Recommendations from the comparison between the CTM systems

The applied CTM modelling systems originate from different institutes and represent independent lines of development. Their operations require varying degrees of the user experience, input data requirements, and computational demand. All three systems are open source, installed and used in a number of countries and possess long records of operational and research applications. The EMEP and SILAM models are usually less demanding than CMAQ in terms of computational resources and input data. Yearly totals of anthropogenic emissions can be used as input to the two models, which perform the temporal disaggregation of emissions in-line with the computation. CMAQ has probably the most-extensive user community with support provided by the developers from the US Environmental Protection Agency (Otte and Pleim, 2010).

Comparison of the model performances does not give an unequivocal answer: all model skills are within the uncertainty of the corresponding parameters (Fig. 3–6). One should however bear in mind that EMEP model was run with lower resolution than two other models. There are certain systematic differences between model results from SILAM and CMAQ on one side and EMEP on the other: e.g. higher NO₂ concentrations at rural stations and lower at urban stations (compare the spatial correlation of annual station averages in Fig. 7). To a large part, this mismatch can be attributed to the spatial distribution of anthropogenic emissions in the SMOKE inventory, applied in the two CTM systems - in comparison with ECLIPSE emissions used by EMEP.

The EMEP/MSC-W model is routinely used for multi-year calculations, facilitating its use for the HELCOM (Baltic Marine Environment Protection Commission - Helsinki Commission) evaluation of trends in the deposition of nitrogen and sulphur in the Baltic Sea region. CMAQ is being used for a variety of environmental modelling problems including regulatory applications and evaluation of emission control strategies (Otte and Pleim, 2010). CMAQ is continuously updated to remain a state-of-the-science regional CTM. A specific advantage of SILAM is the online computation of wildfire emissions and operational input of hourly STEAM ship emissions. The evaluation of modelled daily mean PM_{2.5} showed that RMSE station values for SILAM are within a smaller range (between lower and upper quartile) than the other two models (Fig. 6). The model was also recently applied to 35-years-long global-to-local reanalysis of air quality by Kukkonen et al. (2018). SILAM can therefore be recommended for use in advanced research applications, specifically addressing the abundance and composition of particulate matter.

3.4 Comparison of the ship contribution in the three CTMs

The influence of shipping emissions on the air quality was evaluated for the annual mean concentrations of the three CTMs. The results for the impact of shipping emissions were calculated as differences between the “base” and the “noship” simulations. Results for the absolute ship-related concentrations of O₃, NO₂, SO₂ and PM_{2.5} are shown in Figure 89, the resulting relative ship contribution to annual mean concentrations is shown in Fig. 86-83 and the spatial average of the relative ship contribution is given in Table 53.

3.4.1 Ship contribution to annual mean NO₂

The ship-related annual mean NO₂ concentrations from the three CTMs are in the range of 3–5 µg m⁻³ along the main ship routes. The NO₂ ship contribution decreases to Baltic Sea background values (about 1 µg m⁻³) within a few hundred kilometres

distance from the centre of the shipping routes. Ships emit NO_x mainly in the form of NO, which is however quickly converted to NO₂, thus atmospheric NO_x is mainly in the form of NO₂. The relative contribution of ship emissions to annual mean NO₂ is more than 40 % over the Baltic Sea (Fig. S6) and S3), 22–28 % for the entire Baltic Sea region and 16–20 % in the coastal land areas (Table 5)–3). In particular, NO_x emissions from shipping affect the harbour cities of the region and coastal areas in southern Sweden. ~~SILAM predicts the weakest relative ship influence in the Baltic Proper and the Gulf of Finland. Such local differences~~ Local differences between the models might be due to the different meteorological drivers or differences in the titration efficiency for ozone.

3.4.2 Ship contribution to annual mean O₃

In the proximity of the main shipping routes, negative concentration differences for the modelled annual mean O₃ between the “base” and the “noship” simulation are obtained as a result of the titration effect by the NO_x emissions from shipping. The highest ozone reduction due to shipping is found in the western part of the Baltic Sea. In the CMAQ simulation the depletion of ozone is stronger than in the other two models; with O₃ reduction by 6–12 μg m⁻³ in the Kattegat and in the Danish Straits. The hourly variation of ship emissions is represented in the simulations with CMAQ and SILAM, whereas monthly averaged ship emissions are used in the EMEP/MSC-W simulation. Emission peaks of NO_x from ships that are present in the hourly data can result in occasional stronger ozone titration leading to overall higher reduction of ozone, than it is the case for monthly averaged ship emissions. ~~The SILAM simulation shows only a small influence of shipping~~ Over the coastal land areas, the average impact on annual mean of O₃ along the ship lanes, i.e. the titration effect is much weaker than in the other two models. ~~SILAM shows ozone increases by ca. 2 in a distance of several hundred kilometres from the ship lanes in Baltic Proper, which is not evident for the other models. This might indicate a limited regime in some areas of the Baltic Sea, where ozone increases with increasing from shipping.~~ is very small, with ozone increases between 0.1 % and 0.4 % for the models.

3.4.3 Ship contribution to annual mean SO₂

Ship emissions of SO₂ have a high contribution to annual mean SO₂ concentrations over the Baltic Sea. The ship contribution to SO₂ is 0.5–0.7 μg m⁻³ in a wide corridor around the main shipping routes of the Baltic Sea. While the absolute ship contribution of the three CTMs is similar, the relative ship contribution in the EMEP/MSC-W simulation is higher in most areas of the Baltic Sea and in Sweden, because the background atmospheric SO₂ levels in this simulation are lower than in CMAQ and SILAM.

3.4.4 Ship contribution to annual mean PM_{2.5}

The ship contribution to annual mean PM_{2.5} shows a gradient from southwest to north with highest concentrations over Denmark, the west coast of Sweden, the Belt Sea/Kattegat and over the sea south of Sweden with maximum values up to 0.9 μg m⁻³. The relative contribution in these ship-impacted areas is up to 10 %. ~~In the entire Baltic Sea region the average contribution~~ The average ship contribution for the three CTMs is in the range of 4.34, 1–6.5 % for the three CTMs in the entire Baltic Sea region and 3.1–5.7 % in the coastal land areas. The absolute ship contribution in SILAM is slightly smaller than for the other

two models, in particular in the southwest part of the Baltic Sea region (Fig. 8d) ~~9d~~. ~~A possible explanation for this is the lower dry deposition of fine aerosols in SILAM, in particular over seawater surfaces (Kouznetsov and Sofiev, 2012).~~ The ship-related PM_{2.5} affects the coastal areas in the Baltic Sea region, as its influence extends further inland than it is the case for ship-related NO₂ or SO₂. This can be attributed to the formation of secondary particulate matter in the ship exhaust plume during its transport away from the main shipping routes.

3.5 Comparison of PM_{2.5} in summer and autumn

CMAQ and EMEP/MSC-W simulations predict higher seasonal mean concentrations of PM_{2.5} in autumn (average of September, October and November, SON) than in summer (average of June, July and August, JJA), whereas the SILAM simulation predicts higher PM_{2.5} in summer (Fig. 9 ~~10a,c~~; Table S9 ~~S10~~). The temporal correlation between model data and observations of daily mean PM_{2.5} for the average of the AirBase stations is slightly better in autumn than in summer (Table S7 ~~S8~~ and S8 ~~S9~~). Observed PM_{2.5} in summer is underestimated strongly by CMAQ (at ~~all stations almost all stations~~; see Sect. 3.1.2) and to some extent by EMEP/MSC-W. In autumn all models are in better agreement with observed PM_{2.5}.

The SOA formation mechanism in the applied version of CMAQ (i.e. v5.0.1) is probably not adequate for reproducing the summertime aerosol. Primary organic aerosol (POA), SOA and organic vapours in the atmosphere should be considered a dynamic system that constantly evolves due to multi-generation oxidation (Robinson et al., 2007). We note that multi-generational aging chemistry for the semi-volatile POA was introduced in CMAQ v5.2, based on the approach of Donahue et al. (2012), which considers the functionalization and fragmentation of organic vapours upon oxidation. In addition, wildfire emissions have not been considered in the simulation with CMAQ. Wildfires emit large quantities of organic material and are associated with high biogenic VOC emissions due to high temperature, leading to increased SOA formation (Lee et al., 2008).

In summer, modelled mean PM_{2.5} in the region is much higher in the SILAM simulation (5–8 μg m⁻³ in most parts; 7.4 μg m⁻³ on ~~domain-average regional average~~; 5.4 μg m⁻³ on average in coastal land areas) than for the other two models (< 4 μg m⁻³, except for the urban areas). The higher summertime PM_{2.5} in SILAM is most likely due to more efficient SOA production and ~~different~~ primary emission from wildfires and/or mineral dust. A previous comparison of the models to PM_{2.5} observations from the EMEP station network in Europe reported similar seasonal mean concentrations of the SIA components, i.e. nitrate (NO₃⁻), ammonium (NH₄⁺) and SO₄²⁻, for the three CTMs in summer (Prank et al., 2016).

The calculated ship contribution from all models is higher in summer than in autumn (Table S9 ~~S10~~). The simulations reflect the greater importance of shipping activities during summer and their influence on PM_{2.5} levels over the entire Baltic Sea and the coastal areas (Fig. 9 ~~10b~~). In particular Denmark and the Swedish west coast is highly impacted in summer, with a ship contribution of 0.5–0.9 μg m⁻³ to ambient PM_{2.5} levels.

In autumn, all CTMs predict high levels of PM_{2.5} in the southern part of the Baltic Sea region, exceeding 6 μg m⁻³. High PM_{2.5} in autumn is typically attributed to stagnant meteorological conditions and higher emissions of primary particulate matter from residential heating and energy production. Modelled PM_{2.5} in Sweden and Finland is higher in SILAM than in the other two models. SILAM overestimates observed PM_{2.5} at the stations in Sweden, Lithuania and Finland in summer (NMB: 0.54 on

average) and autumn (NMB: 0.31 on average). In an earlier model comparison, all three models were shown to overestimate NO_3^- and NH_4^+ in autumn; while SILAM also overestimated SO_4^{2-} in autumn (Prank et al., 2016) (Prank et al., 2016).

The ship contribution in autumn in the southwest part of the region is higher in EMEP/MSC-W compared to the other two models (Fig. 910d), obviously a result from larger secondary formation of particulate matter, as mainly the coastal regions are impacted. The formation of SIA in autumn is favoured by lower temperature and higher humidity compared to summer. ~~Since The higher autumn ship contribution in the EMEP model can be attributed to differences in land-based anthropogenic emissions of NH_3 and NO_2 as well as the ship emissions are the same in CMAQ and SILAM, the lower autumn ship contribution in SILAM is mainly attributed or~~ to differences in the schemes for inorganic aerosol formation. The investigation of differences between the SIA formation schemes is, however, beyond the scope of this study.

10 3.6 Comparison of elemental carbon related to ship emissions

Primary carbonaceous particles emitted from ships are the product of incomplete fuel combustion and consist of a mixture of elemental carbon and non-polar organic carbon. In the STEAM ship emission inventory these are separated into emissions of elemental carbon (EC) and organic carbon (OC). The terms EC and BC are used interchangeably in the models, however both can only be regarded as proxies for the concentration of soot particles (Vignati et al., 2010). Here we are mainly interested in the atmospheric fate of EC from ship emissions, as simulated by the models. EC particles are associated with adverse human health effects (Dockery et al., 1993) and contribute to regional haze and poor visibility (e.g. Odman et al., 2007). The atmospheric lifetime of EC is relatively long; around 6 days in the continental outflow (Park et al., 2005) and 4–8 days on global scale (Vignati et al., 2010), with a large uncertainty due to soot ageing processes and wet deposition (Textor et al., 2006). Therefore the removal by deposition within the study region is expected to be rather limited.

20 The spatial averages of the EC concentrations (“base” simulation) and the ship-contributed EC concentrations are given in Table 64. The seasonality of ship-related EC predicted by the three CTMs is shown in Fig. 4011. The levels of ship-related EC are higher in spring and summer than in autumn and winter due to more intense shipping activities. Therefore the ship contribution peaks in the seasons when ambient EC concentrations are lowest (Table 64; Fig. S7S4). The highest levels of ship-related EC, in the range of 0.03–0.04 $\mu\text{g m}^{-3}$, occur along the main shipping routes and in the main ports of the region. ~~On regional average, ships contribute 5–7% to the~~ The average ship contribution to annual mean EC is 4–5% over coastal land regions (Table 5)-3).

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 $\mu\text{g m}^{-3}$ 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 $\mu\text{g m}^{-3}$; SILAM: 0.0144 $\mu\text{g m}^{-3}$, EMEP/MSC-W: 0.0149 $\mu\text{g m}^{-3}$). 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.

SILAM predicts a stronger seasonal variability of the ship-related EC than the other models. In particular, modelled EC ship contribution ~~is spring and winter are in winter is~~ lower than for CMAQ and EMEP/MSC-W.

Shipping emissions of EC are identical in the three CTMs on a monthly basis. Differences between the models are therefore explained by differences in the meteorological conditions, ~~deposition schemes and~~ the treatment of atmospheric transport in the models ~~and the vertical ship emission profiles. In CMAQ and EMEP/MSC-W all ship emissions are injected below 100 height. The lower surface concentrations of ship-related in EMEP/MSC-W can thus be attributed to different atmospheric stability, i.e. more frequent neutral conditions, which dilutes the concentration of the emitted pollutant. The fact that is more confined to the shipping routes and shows a limited spatial spreading compared to the other models might also indicate less efficient horizontal diffusion in EMEP/MSC-W.~~

~~In SILAM, a vertical ship emission profile that specifically distributes the STEAM emissions from 0 up to 1000 above the ground, to mimic the plume rise of ship exhaust (Table 2). This way, typically 38 % of the ship emissions are injected in a height above 500 in the model. In stable conditions, the boundary layer (BL) height over the Baltic Sea is often at or below 500 m (Svensson et al., 2016; Gryning and Batchvarova, 2002), which would effectively prevent pollutant mass emitted in higher vertical layers to reach the surface layer.~~ Climatological simulations over the Baltic Sea show that there is a strong seasonality in the atmospheric stability over the sea with more than 50 % stable conditions in spring whereas during the other seasons unstable conditions dominate together with occasionally neutral conditions (Svensson et al., 2016). ~~Stable conditions over the Baltic Sea in the SILAM simulation will reduce the quantity of ship emissions that can affect the surface layer concentrations. Thus in SILAM a larger proportion of the ship emissions becomes subject to long-range transport and is exported out of the Baltic Sea region~~ The lower surface concentrations of ship-related EC in SILAM and EMEP/MSC-W during spring compared to CMAQ might be attributed to different atmospheric stability in the simulations, i.e. more frequent occurrence of neutral conditions, which dilutes the concentration of the emitted pollutant. The fact that ship-related EC in EMEP/MSC-W is more confined to the shipping routes and shows a limited spatial spreading compared to the other models might also indicate less efficient horizontal diffusion. In winter, SILAM and EMEP/MSC-W ship-related EC is very low and the impacted area has a smaller extent than in the other months (white areas in Fig. 4011), indicating faster removal of EC particles than in the CMAQ simulation. Different treatment of the hygroscopicity and ageing processes of EC particles, affecting their wet scavenging, could have contributed to the differences among the models.

3.7 Comparison of oxidised nitrogen deposition

~~Annual atmospheric deposition of total nitrogen to the Baltic Sea basin has declined by about one third between 1995 (305) and 2015 (222) (Bartnicki et al., 2017). During the two decades, the deposition of oxidised nitrogen (NO_x) decreased by 35 % whereas the deposition of reduced nitrogen (NH_3), i.e. and NH_4^+ , decreased by only 12 % (Bartnicki et al., 2017).~~

~~Results of the spatial distribution of oxidised nitrogen deposition from the three models were compared (Fig. S8). The annual deposition of NO_x shows a strong gradient from southwest to northeast in the simulation by all models. Deposition of NO_x to the Baltic Proper is larger in SILAM and EMEP/MSC-W (280–350) than in CMAQ (230–290). The companion study by Karl et al. (2018) reports a wide-spread underestimation of the wet deposition of nitrate and ammonium by CMAQ in~~

2012 compared to observations of the regional background monitoring stations of the EMEP network. The underestimation of nitrogen deposition in CMAQ is likely caused by a combination of too slow oxidative conversion of NO_x to NO_3^- , low particulate ammonium from the regional background and underestimation of precipitation in the southern Baltic Sea (Karl et al., 2018).

The ship-related annual deposition of oxidised nitrogen is however similar among the models and on average 40–60 over the Baltic Sea. The relative contribution of shipping emissions to the deposition of NO_3^- to the Baltic Sea is 24% for CMAQ (?) and 14–18% for the other two models. The contribution from ships to the oxidised nitrogen deposition in the Baltic Sea basin for year 2012 in the present study is therefore higher than the previous reported range from 12 to 14% (Hongisto, 2014) for the period 2008 to 2011. The ship-related contribution to the nitrogen deposition to the Baltic Sea is in the range of 21–23 for the three CTMs of this study (Fig. 11), thus 40–70% higher than for single years of the Hongisto (2014) study. The higher ship contribution in the present study might partially be due to the inter-annual variation of nitrogen deposition caused by changing meteorological conditions, which is typically in the range of 13% to 17% (Hongisto, 2014). Most of northern Europe experienced above-average precipitation during 2012; in Finland the annual precipitation in 2012 was 739, which is 173 above the 1961–1990 average (WMO, 2013). The Hongisto (2014) study applied the STEAM ship emission inventory and fine resolution grids (0.068), however the specific treatment of formation or the abundance of atmospheric oxidants and represent possible explanations for the discrepancy.

The total nitrogen deposition to the Baltic Sea when all emissions are considered is ~230 for EMEP/MS-CW and SILAM, comparable to the estimate of 223.6 (2012; normalised by inter-annual changes in meteorological conditions) used in the HELCOM evaluation of the Baltic Sea marine environmental status (Bartnicki et al., 2017) which uses the EMEP model with the coarser 50-resolution. CMAQ computes lower deposition totals: is 25% lower and is 40% lower compared to the other two models (Fig. 11). According to the SILAM simulation, wet deposition has a share of 60% and 66%, respectively, to the total deposition of NO_3^- to the Baltic Sea. The wet deposition share was similar in the EMEP simulation but lower in the CMAQ simulation (54%; 63%). For SILAM and EMEP/MS-CW, the wet deposition share to total nitrogen deposition is within the range of 63–70% given for the years 1995–2006 (Bartnicki et al., 2011).

4 Summary and conclusions

The effect of ship emissions on the regional air quality and nitrogen deposition in the Baltic Sea region was investigated with three regional CTM systems (CMAQ, ~~EMEP/MS-CW~~ EMEP/MS-CW and SILAM) that simulate the transport, chemical transformation and fate of atmospheric pollutants. The models were applied with their typical setup for air quality studies in European regions. ~~The use of three different CTM systems has the advantage of providing the necessary information for assessing the uncertainties connected with the atmospheric transport and transformation of pollutants.~~ The same ship emission dataset from the STEAM model based on ship movements from AIS records, detailed ship characteristics and up-to-date load dependent emission factors were used in all CTMs. The models were set up with a finer grid resolution (4-km to 11-km grid length) than it was the case for previous air quality studies in the Baltic Sea region, potentially enabling a better treatment of the dispersion and photochemistry in exhaust plumes from shipping along the major ship lanes of the Baltic Sea.

The comparison of air total concentrations of regulatory air pollutants among the models is the primary focus of this study. Results from the three CTMs were compared to observations from rural and urban background stations of the AirBase monitoring network in the coastal areas of the Baltic Sea region. ~~The performance of the models to predict pollutant concentrations was found to be similar.~~ The finer resolution of CMAQ and SILAM (4 km) compared to ~~EMEP/MSC-W~~ EMEP/MSC-W (11 km) did not lead to a significant improvement of the urban bias and urban temporal correlation for daily mean NO₂ concentrations. The benefit from using high-resolution grids depends on the availability of accurate urban emission data with high spatial resolution (Schaap et al., 2015) and realistic temporal profiles (Kukkonen et al., 2012). While the STEAM inventory provides this data for shipping, the compilation of urban emission inventories is more challenging because they are based on specific information for each sector, such as housing units for domestic heating or number of vehicles (Guevara et al, 2016). Observed PM_{2.5} in summer is underestimated strongly by CMAQ at all stations and to some extent by ~~EMEP/MSC-W~~ EMEP/MSC-W. Observed PM_{2.5} in winter is underestimated by SILAM. In autumn all models are in better agreement with observed PM_{2.5}. The low summer PM_{2.5} in CMAQ has been attributed to the underestimation of secondary organic aerosols and to the missing emissions of wind-blown soil dust particles (~~Karl et al., 2018~~) (Karl et al., 2019). Particulate matter emissions from wind-blown dust and forest fires ~~are included in EMEP/MSC-W~~ were included in EMEP/MSC-W and SILAM.

~~The ship influence on concentrations~~ Ship-related concentrations of NO₂ were evaluated at coastal monitoring stations ~~was investigated with a statistical method. A station is regarded as ship-influenced if the modelled concentration as well as the correlation with observed increased significantly (on 0.9 level or higher) by testing if the agreement between predicted and observed total concentrations improves significantly~~ when ship emissions are included in the ~~CTM simulation.~~ simulation. Including ship emissions improved the agreement between modelled and measured NO₂ daily mean concentrations at about 50 % of the stations. The change of modelled O₃ due to shipping was below 6 % on summer average at the shoreline stations and mainly reflects additional ozone production due to ship emissions. Ship-influenced stations identified by the models are mainly located close to the shoreline or close to a port; ~~but a few inland stations up to 200 distant from the shore were also found to be influenced from shipping, with a maximum distance of 20 km from the sea.~~ However, modelled peaks of high daily mean NO₂ at coastal rural sites during summer that are not present in the measurements indicate that the models often did not properly resolve the ship plumes due to the sub-grid variability of the plume dispersion of individual ships.

The spatial average of annual mean O₃ concentrations in the ~~EMEP/MSC-W simulation is 15–25~~ EMEP/MSC-W simulation is ca. 20 % higher compared to the other two simulations. ~~EMEP/MSC-W~~ EMEP/MSC-W overestimates the measurements of daily mean O₃ concentrations at rural stations by 17 % on average. The higher ozone concentrations in the EMEP model are mainly the consequence of using a different set of boundary conditions for the European model domain. The concentration gradients of NO₂ and SO₂ between urban and rural areas and between ship lanes and the surrounding sea are larger in ~~EMEP/MSC-W~~ EMEP/MSC-W than in the other models. Factors contributing to the different gradients are differences in the representation of horizontal transport, differences in the meteorological driving data, and differences in the atmospheric lifetime of NO₂ and SO₂ in the models.

There are significant differences in the calculated ship contributions to the levels of air pollutants among the three models. In the proximity of the main shipping routes, ozone is depleted as a result of the titration effect by NO emissions from shipping.

~~SILAM predicted a much weaker titration effect than Ozone depletion in EMEP/MSC-W is weaker than in the other two models. In contrast with the other models, SILAM shows ozone increases due to shipping several hundred kilometres away from the shipping lanes in the Baltic Proper, due to a combination of the larger grid cell volume causing higher initial dilution of ship emissions and of the use of monthly averages for ship emissions, averaging out hourly emission peaks.~~

5 The ship-related PM_{2.5} affects the coastal areas in the Baltic Sea region, as its influence extends further inland than it is the case for ship-related NO₂ and SO₂. ~~In the entire Baltic Sea region the~~ The average contribution of ships to levels of PM_{2.5} is in the range of 4.3 ~~calculated by the three CTMs ranges between 4.1–6.5 % for the three CTMs in the entire Baltic Sea region and between 3.1–5.7 % in the coastal land areas.~~ Differences in ship-related PM_{2.5} among the models are mainly attributed to differences in the schemes for inorganic aerosol formation.

10 Since shipping emissions of elemental carbon are identical in the three CTMs on a monthly basis, differences for ship-related EC can be explained by differences in the meteorological conditions, the treatment of atmospheric transport ~~in the models and the vertical ship emission profiles. In SILAM a large proportion of the ship emissions is injected in altitudes above 500 and becomes subject to long-range transport out of the Baltic Sea region.~~

~~The ship-related annual deposition of oxidised nitrogen is similar among, and the wet scavenging parameterizations in the~~ models. The annual oxidised nitrogen deposition from ships to the Baltic Sea is on average 40–60 and annual totals are in the range of 21–23, and thus higher than previous estimates. The higher ship contribution in the present study might partly be a result of the high precipitation during 2012.

Results obtained from the use of three CTMs give a more robust estimate of the ship contribution to atmospheric ~~concentrations and deposition~~ pollutant concentrations than a single model. By using several models the sensitivity of the ship contribution to uncertainties of boundary conditions, meteorological data as well as aerosol formation and deposition schemes is taken into account. This is an important step towards a more reliable evaluation of policy options regarding emission regulations for ship traffic and the introduction of a nitrogen emissions control area (NECA) in the Baltic Sea and the North Sea in 2021. ~~In order to improve the calculation of the ship emission influence on local and regional air quality further, better constraints for the vertical emission profile from ships are needed. The plume rise from moving ships should be studied under atmospheric stability conditions relevant in the European seas with the aim to derive generally applicable parameterisations for the vertical profile.~~

Data availability. Data from the simulations with CMAQ, SILAM and EMEP/MSC-W on air pollutant concentrations and nitrogen deposition are available upon request.

Appendix A: Statistical indicators

30 The performance of each model is quantified in terms of mean values (μ_{Mod} and μ_{Obs}), normalized mean bias (NMB), Spearman's correlation coefficient (R), root mean square error (RMSE) and FAC2.

The normalized mean bias is given by:

$$\text{NMB} = \frac{\overline{M} - \overline{O}}{\overline{O}}, \quad (\text{A1})$$

where M and O stand for model and observation results, respectively. The overbars indicate the time average over N time intervals (number of observations). The time average is done for one year, hence \overline{M} corresponds to μ_{Mod} and \overline{O} corresponds to μ_{Obs} .

The Spearman's correlation coefficient is defined as the Pearson's correlation coefficient between the ranked variables. For a sample of size N , the N raw scores M_i and O_i for each time step i are converted to ranks $rg(M_i)$ and $rg(O_i)$. The Spearman's correlation coefficient is then computed from (Myers and Sirois, 2006):

$$R = \frac{\text{cov}(rg(M), rg(O))}{\text{STDR}_M \cdot \text{STDR}_O}, \quad (\text{A2})$$

where $\text{cov}(rg(M), rg(O))$ is the covariance of the rank variables, STDR_M and STDR_O are the standard deviations of the ranks of the model and observation data. The Spearman correlation between two variables is equal to the Pearson correlation between the rank values of those two variables; while Pearson's correlation assesses linear relationships, Spearman's correlation assesses monotonic relationships (whether linear or not). If there are no repeated data values, a perfect Spearman correlation of +1 or -1 occurs when each of the variables is a perfect monotone function of the other.

The root mean squared error combines the magnitudes of the errors in predictions for various times into a single measure and is defined as:

$$\text{RMSE} = \sqrt{\frac{1}{N} \sum_{i=1}^N (M_i - O_i)^2}. \quad (\text{A3})$$

RMSE is a measure of accuracy and allows to compare prediction errors of different models for a particular dataset.

FAC2 is the fraction of model values within factor 2 of the observations (Hanna and Chang, 2012):

$$\text{FAC2} :: \text{fraction where } 0.5 \leq \frac{M_i}{O_i} \leq 2. \quad (\text{A4})$$

Appendix B: [Time series plots](#)

[The appendix contains the time series plots of NO₂ concentrations at selected ship-influenced monitoring stations for the meteorological year 2012. Time series plots of O₃ concentrations at the corresponding sites are shown in Fig. S1.](#)

Author contributions. MK was responsible for the overall structure and did most of the writing, performed simulations with CMAQ, developed a framework for statistical processing, visualization and plotting and evaluated the air concentration data from all models. JEJ performed simulations with the EMEP/MSC-W model and provided model data for the comparison. AU and MP performed SILAM simulations and provided model output. AA carried out the statistical evaluation of the ship contributions and programmed R-scripts for visualization and data processing. MS gave advice on performing the intercomparison. JPJ and LJ provided ship emissions with STEAM. MQ developed research questions, discussed the manuscript structure and assisted with the introduction, conclusions and abstract. VM developed research questions and coordinated the intercomparison. All contributed to the writing of the paper and discussion of the model results.

Competing interests. The authors declare that they have no conflict of interest

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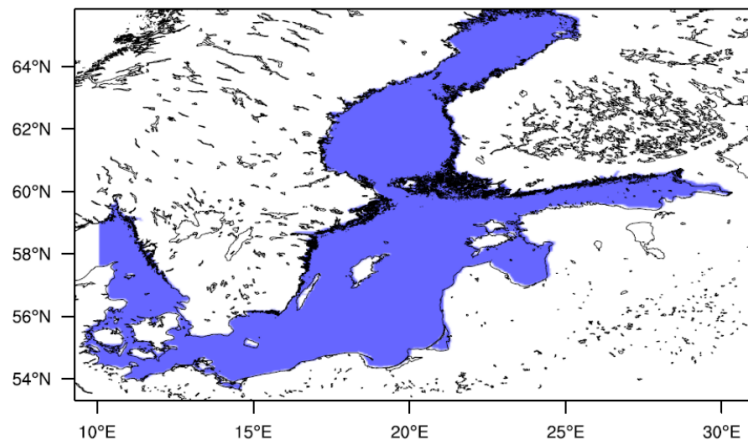


Figure 1. Geographic map of the study domain for the CTM comparison, spanning from latitude 53.40°N (south) to 65.80°N (north) and longitude 9.00°E (west) to 31.10°E (east). The extent of the Baltic Sea as used in this study is shown in blue.

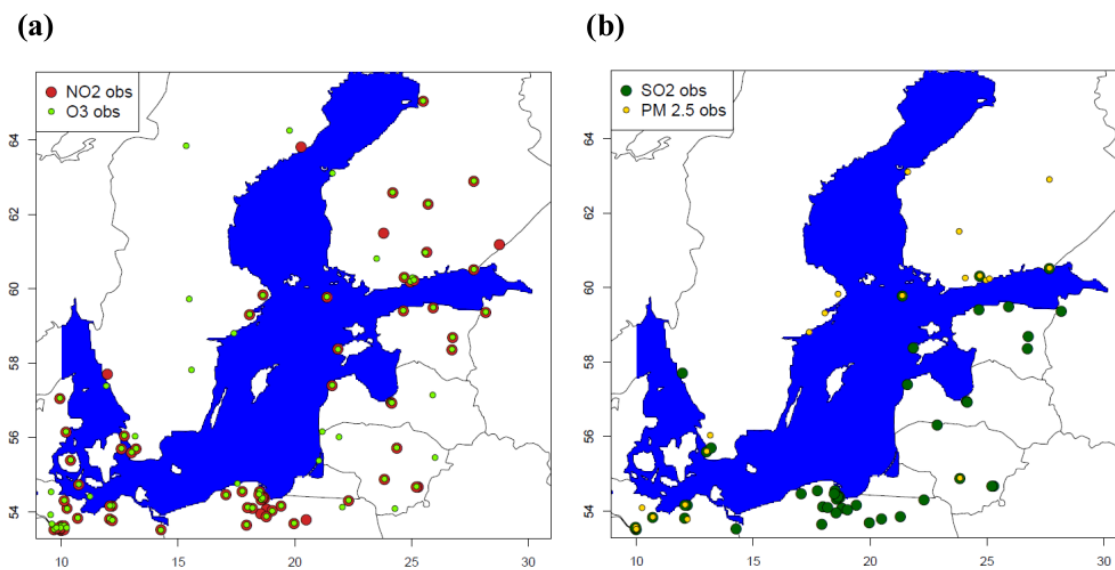
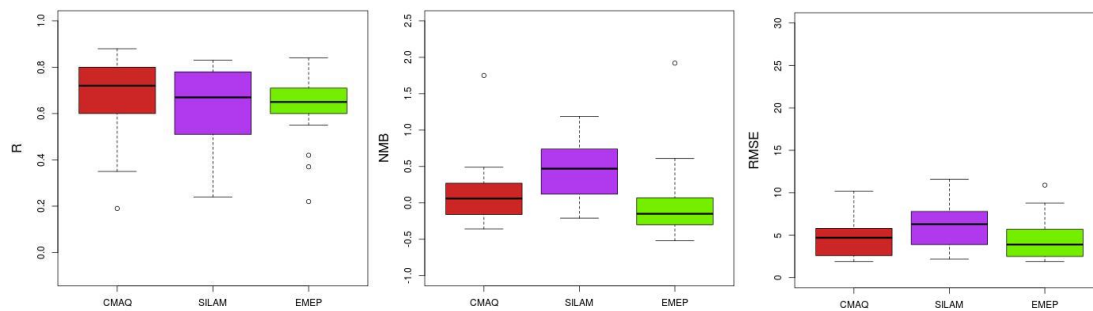


Figure 2. Map of the Baltic Sea region with the location of background monitoring stations used in the statistical performance analysis with observations of: (a) NO₂ (filled red circles) and O₃ (filled green circles); and (b) SO₂ (filled dark green circles) and PM_{2.5} (filled yellow circles). Same domain extent as in Figure 1.

(a)



(b)

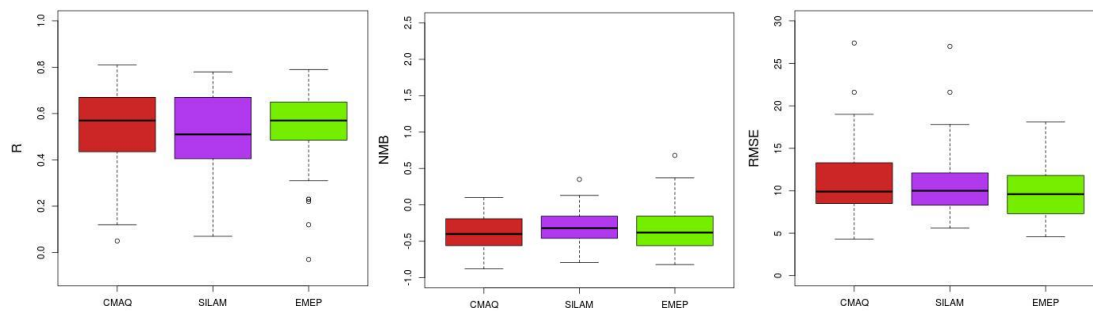
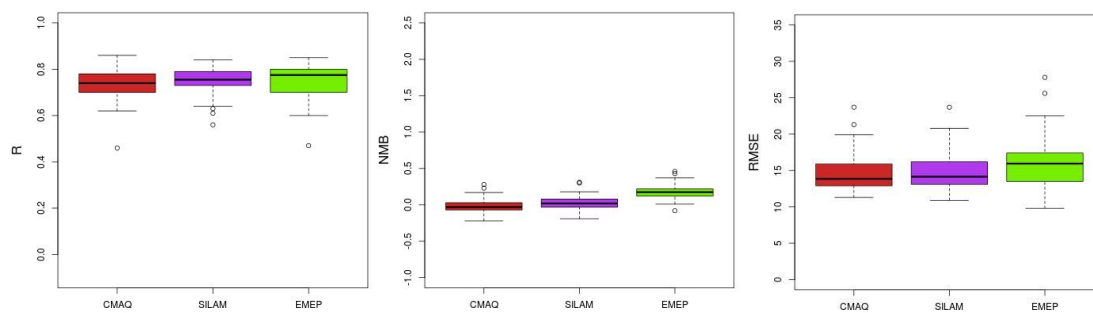


Figure 3. Comparison of statistical indicators for NO₂ daily means (in the order R, NMB, and RMSE) between three CTMs at: (a) rural background stations and (b) urban background stations. Outlier shown as small circles.

(a)



(b)

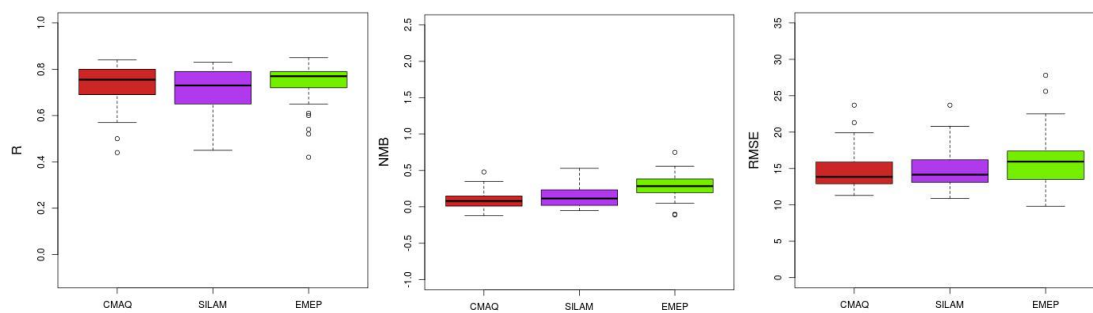
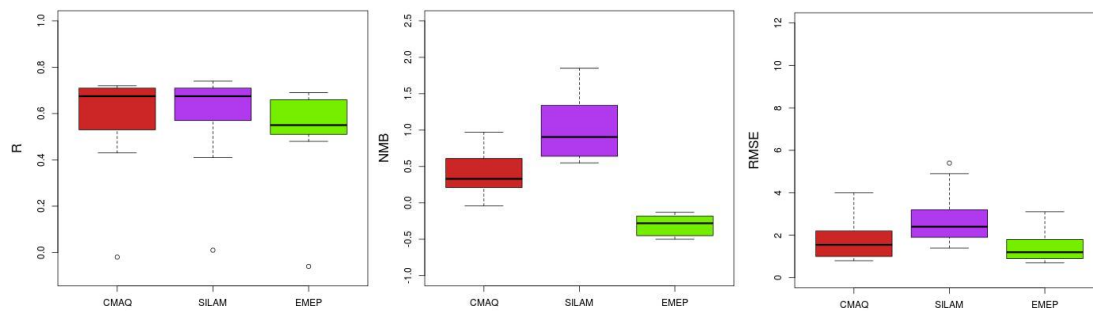


Figure 4. Comparison of statistical indicators for O₃ daily means (in the order R, NMB, and RMSE) between three CTMs at: (a) rural background stations and (b) urban background stations. Outlier shown as small circles.

(a)



(b)

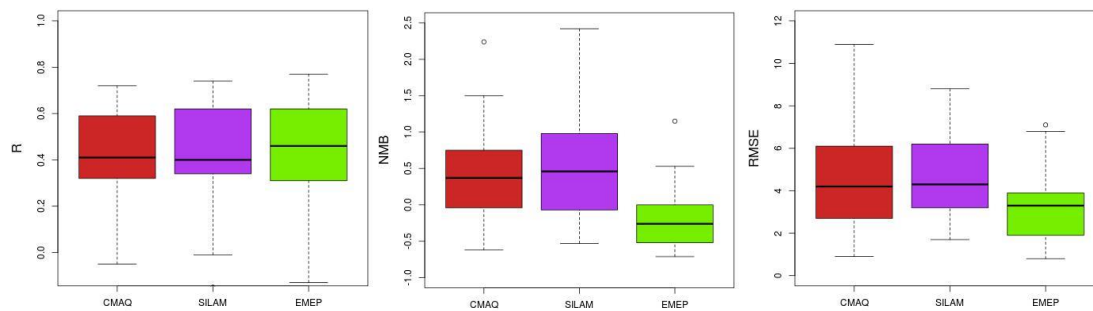
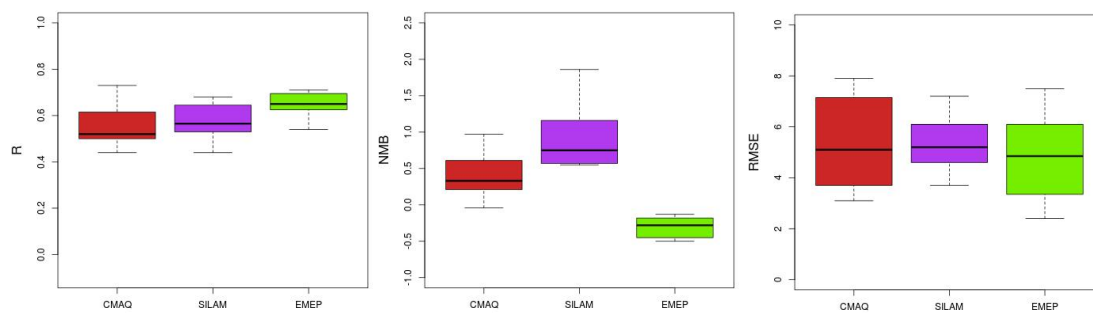


Figure 5. Comparison of statistical indicators for SO₂ daily means (in the order R, NMB, and RMSE) between three CTMs at: (a) rural background stations and (b) urban background stations. Outlier shown as small circles.

(a)



(b)

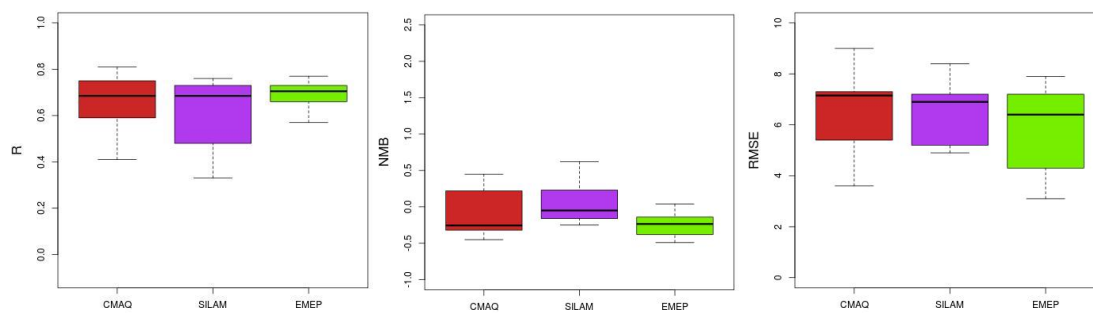


Figure 6. Comparison of statistical indicators for $PM_{2.5}$ daily means (in the order R, NMB, and RMSE) between three CTMs at: (a) rural background stations and (b) urban background stations. Outlier shown as small circles.

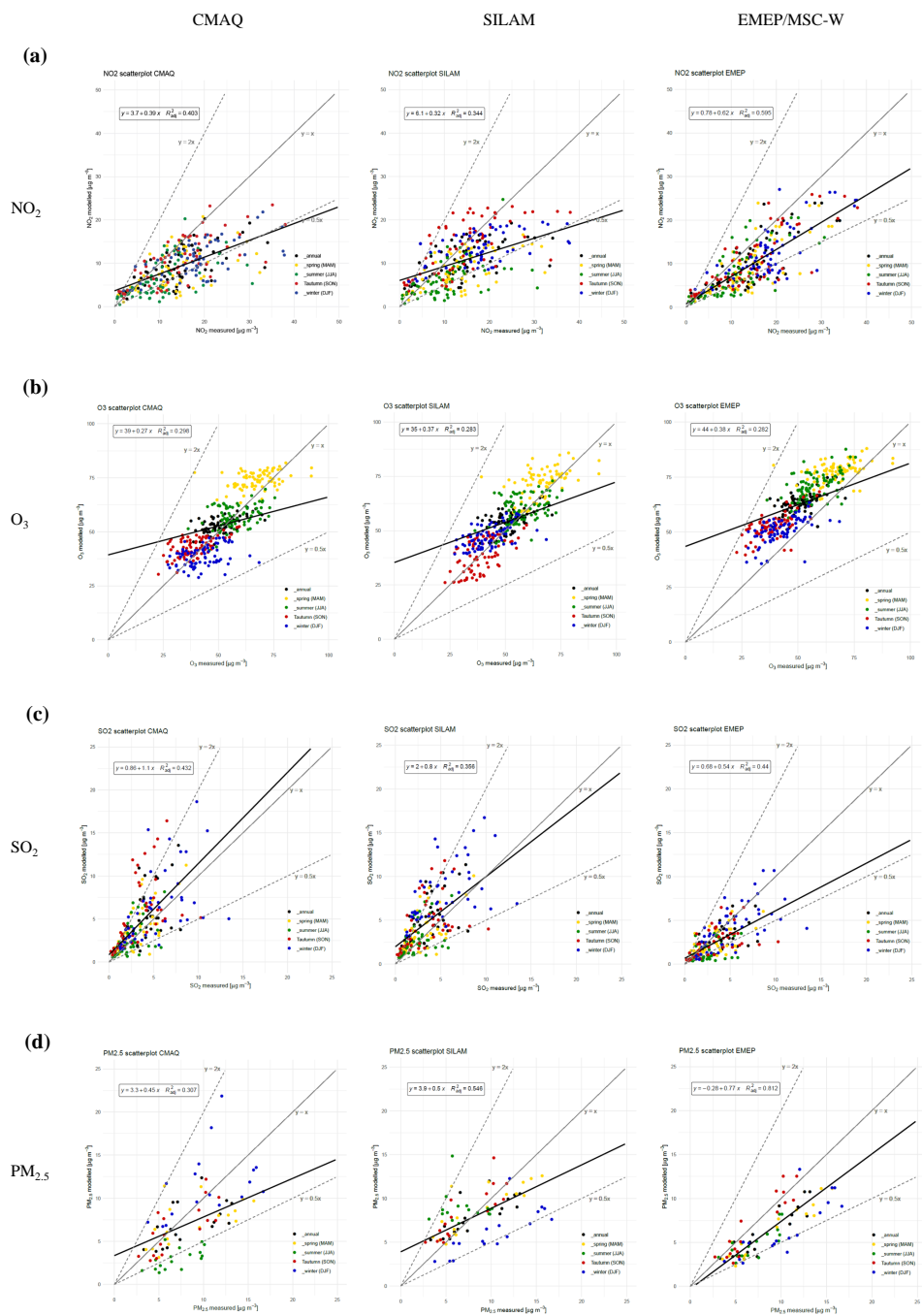


Figure 7. Spatial correlation of annual mean concentrations ($\mu\text{g m}^{-3}$) and the seasonal averages from CMAQ (left column), SILAM (middle column) and EMEP (right column) in the Baltic Sea region for (a) O₃, (b) NO₂, (c) SO₂, and (d) PM_{2.5}.

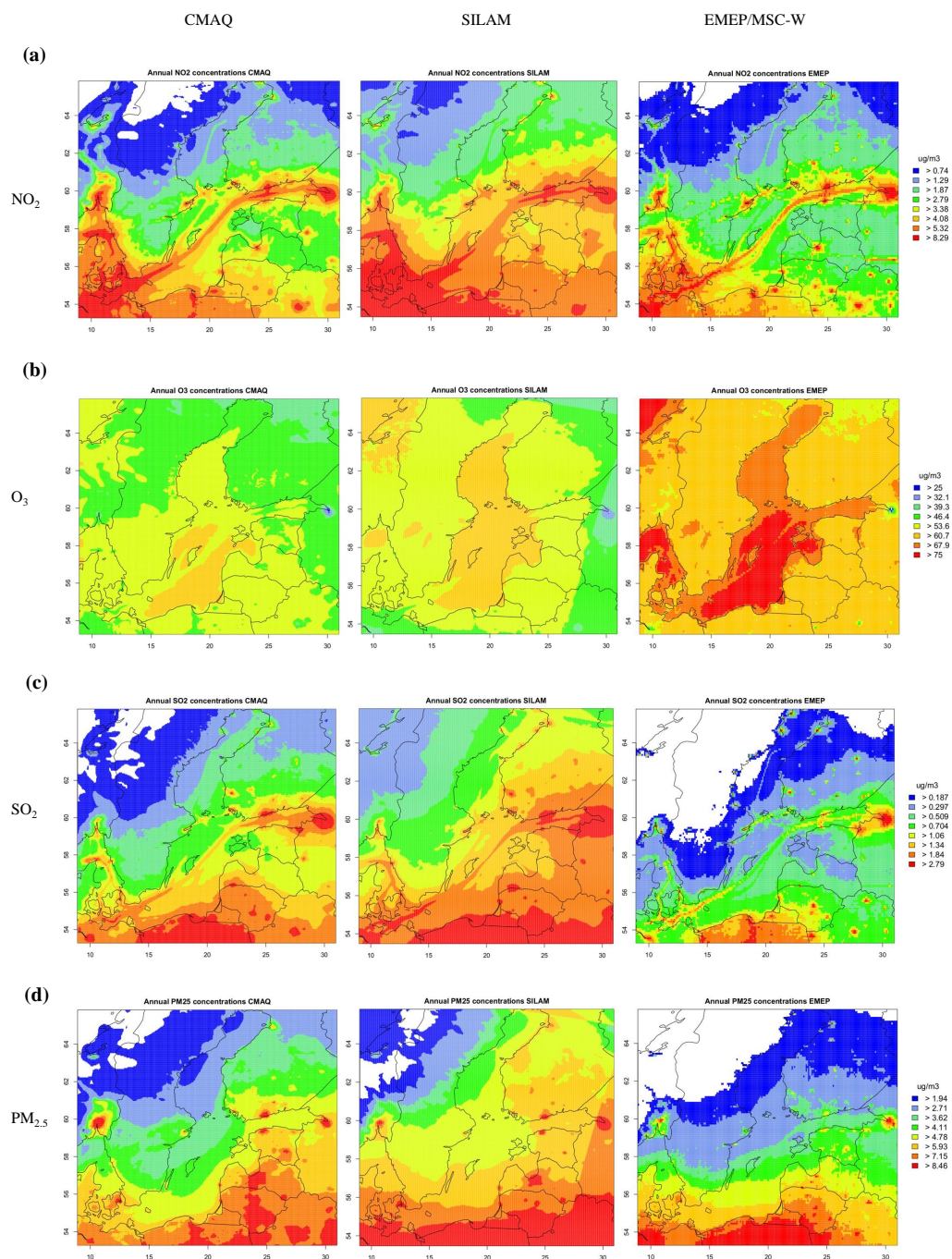


Figure 8. Comparison of the spatial distribution of annual mean concentrations ($\mu\text{g m}^{-3}$) from CMAQ (left column), SILAM (middle column) and EMEP (right column) in the Baltic Sea region for (a) O_3 , (b) NO_2 , (c) SO_2 , and (d) $\text{PM}_{2.5}$. Empty areas correspond to concentrations between zero and the lowest value in the legend.

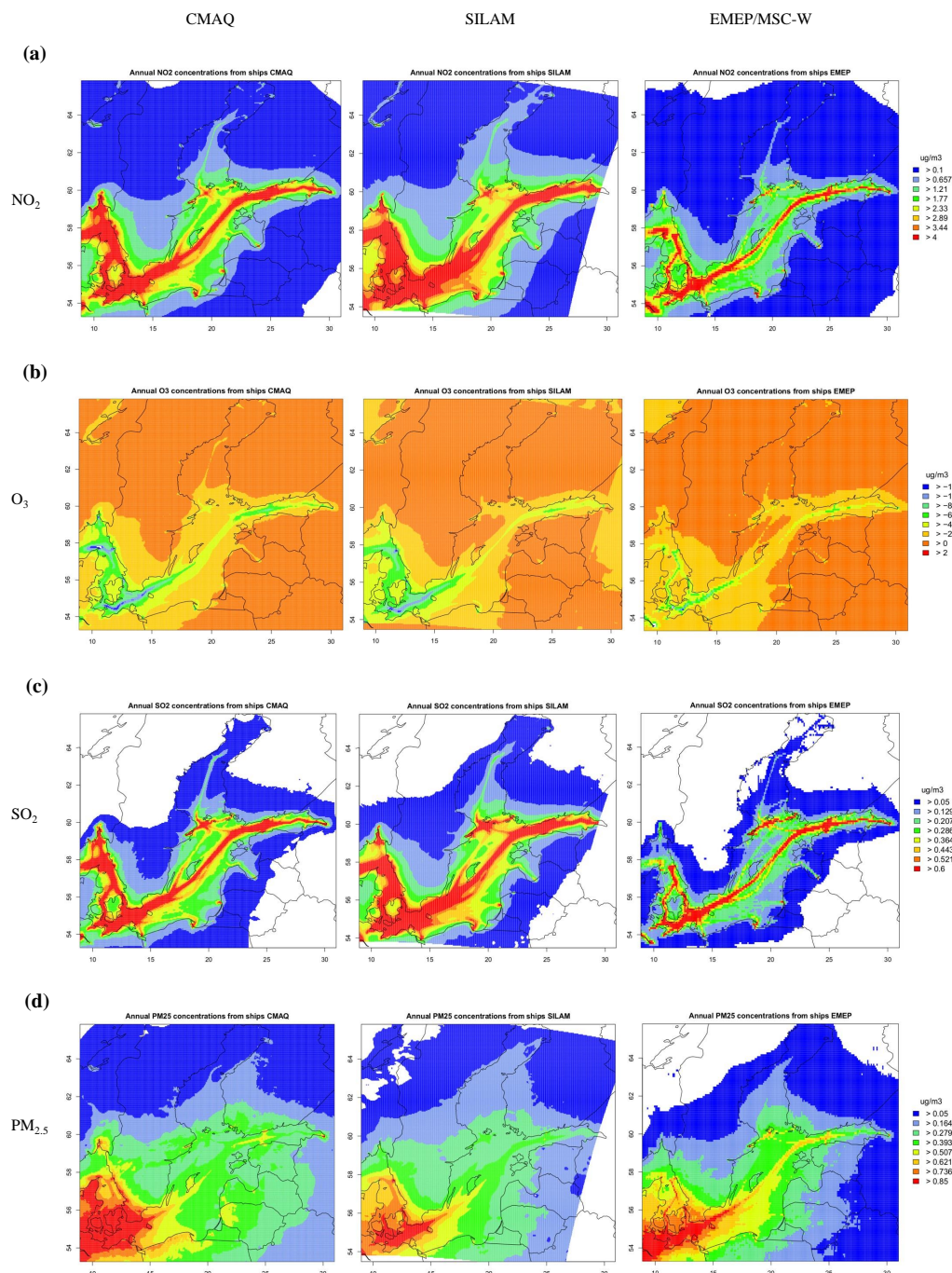


Figure 9. Comparison of the spatial distribution of annual mean ship-related concentrations (absolute ship contributions in $\mu\text{g m}^{-3}$) of the CMAQ (left column), SILAM (middle column) and EMEP (right column) models in the Baltic Sea region for (a) O₃, (b) NO₂, (c) SO₂, and (d) PM_{2.5}.

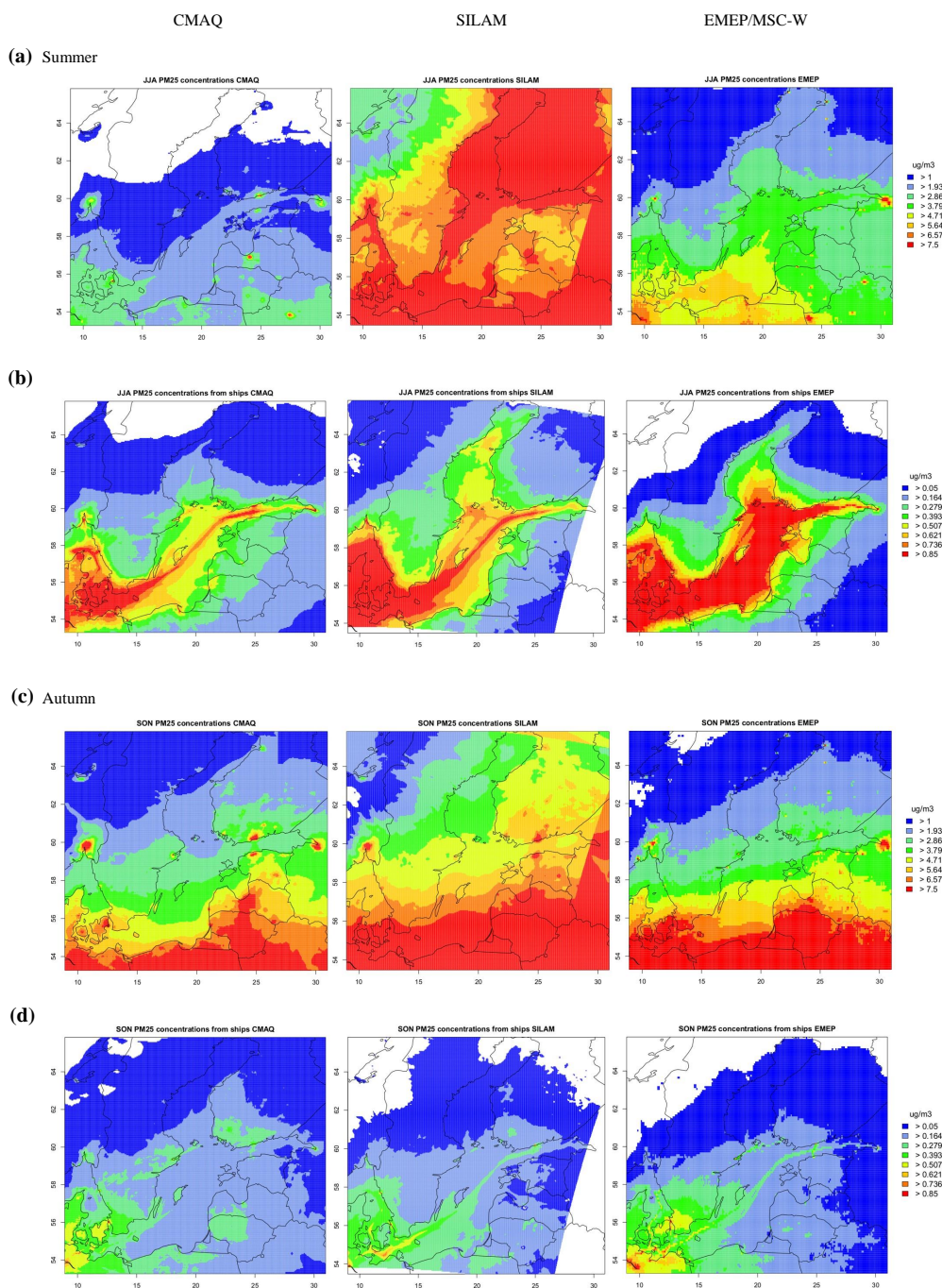


Figure 10. Comparison of PM_{2.5} in summer and autumn from CMAQ (left column), SILAM (middle column) and EMEP (right column) in the Baltic Sea region for (a) JJA mean concentration ($\mu\text{g m}^{-3}$), (b) JJA mean ship contribution ($\mu\text{g m}^{-3}$), (c) SON mean concentration, and (d) SON mean ship contribution. Empty areas correspond to concentrations between zero and the lowest value in the legend.

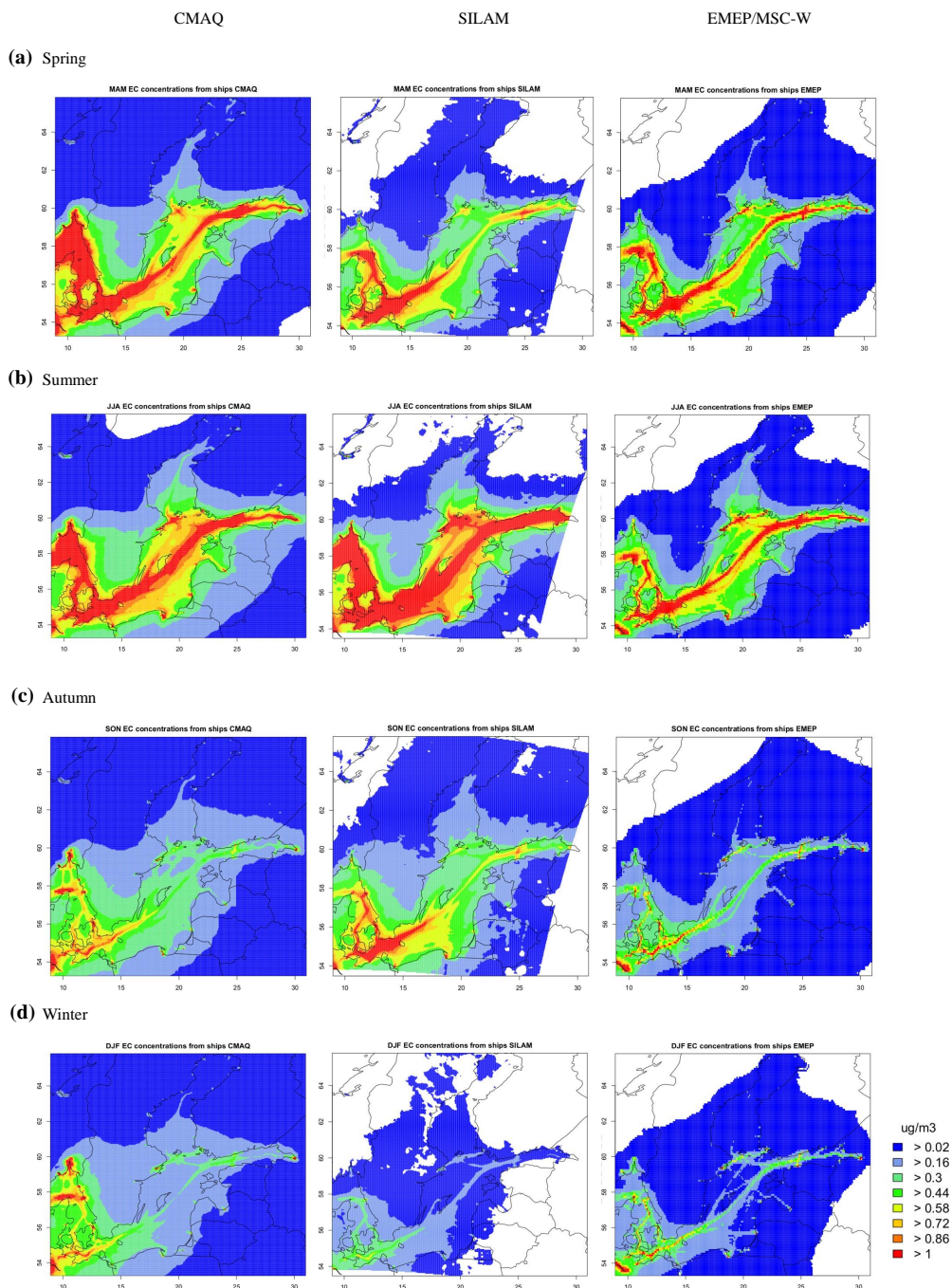


Figure 11. Spatial distribution of the seasonal mean EC ship contribution ($\mu\text{g m}^{-3}$) from CMAQ (left column), SILAM (middle column) and EMEP (right column) in the Baltic Sea region for (a) spring, (b) summer, (c) autumn, and (d) winter. Empty areas correspond to concentrations between zero and the lowest value in the legend.

Figure 11. Comparison of the total annual deposition (ktN y^{-1}) of reduced and oxidised nitrogen and ship-related oxidised nitrogen (N_{ship}) to the Baltic Sea (seawater) from CMAQ (red), SILAM (dark blue) and EMEP/MS-CW (green).

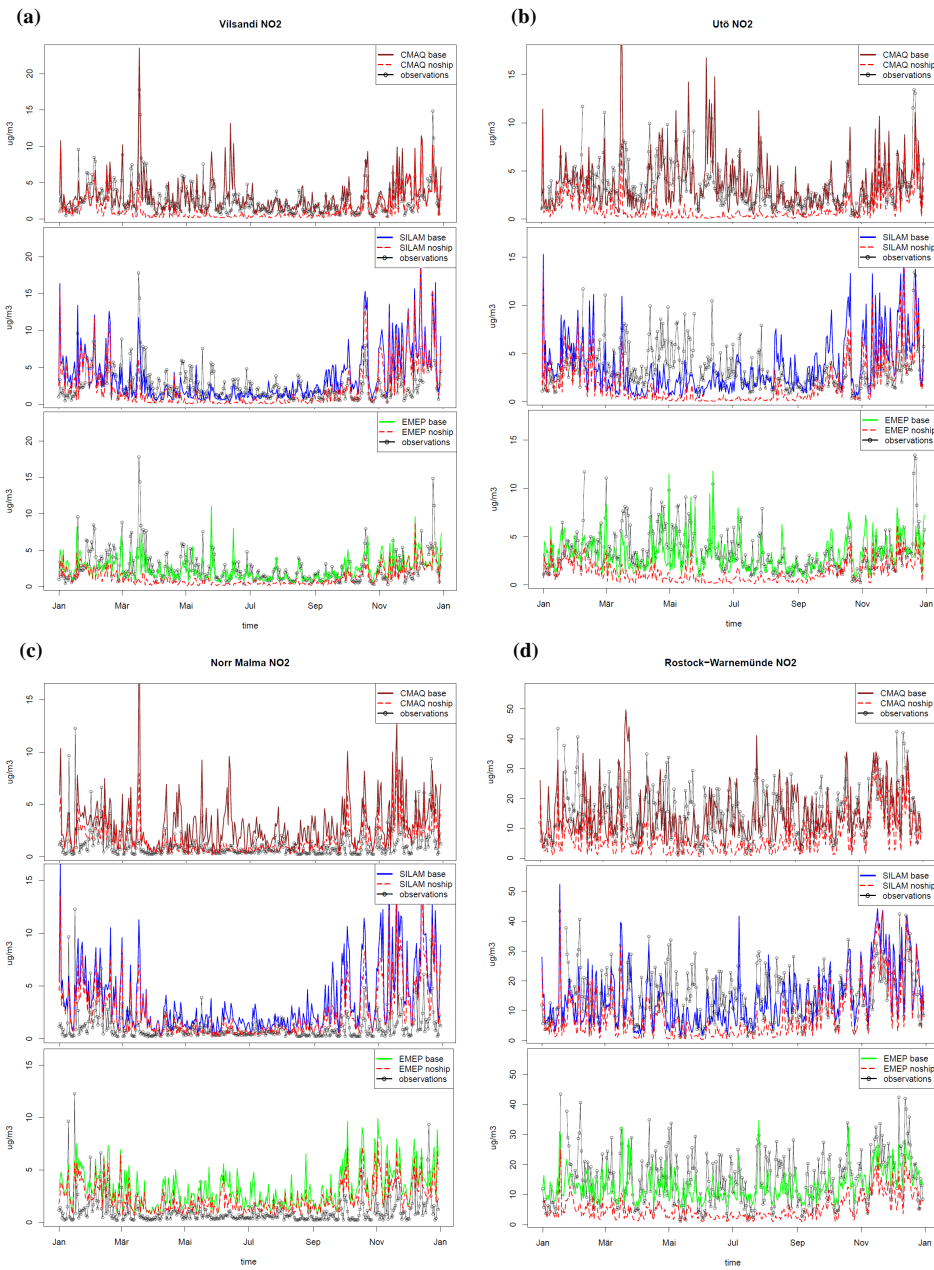


Figure B1. Continued.

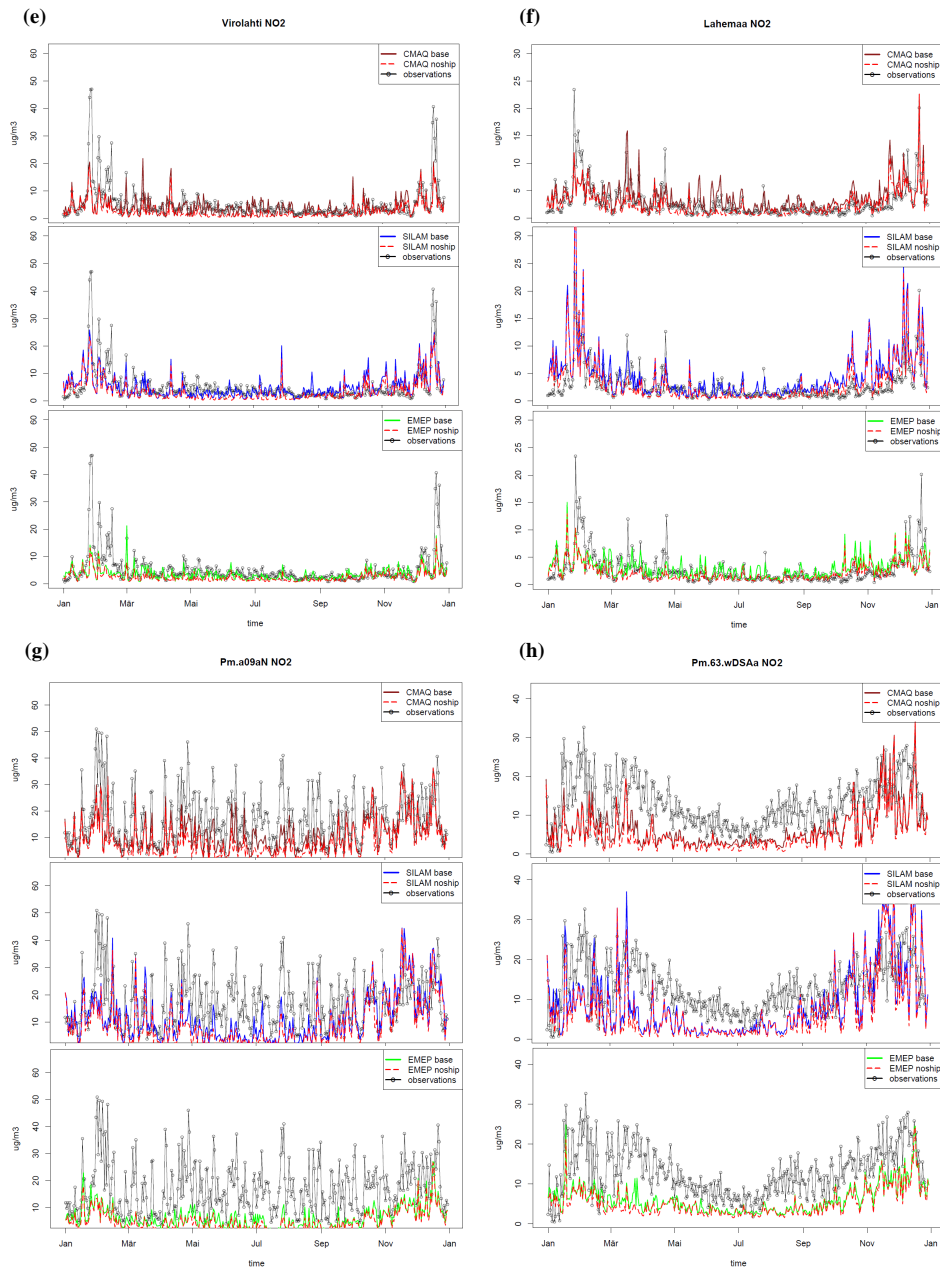


Figure B1. Continued.

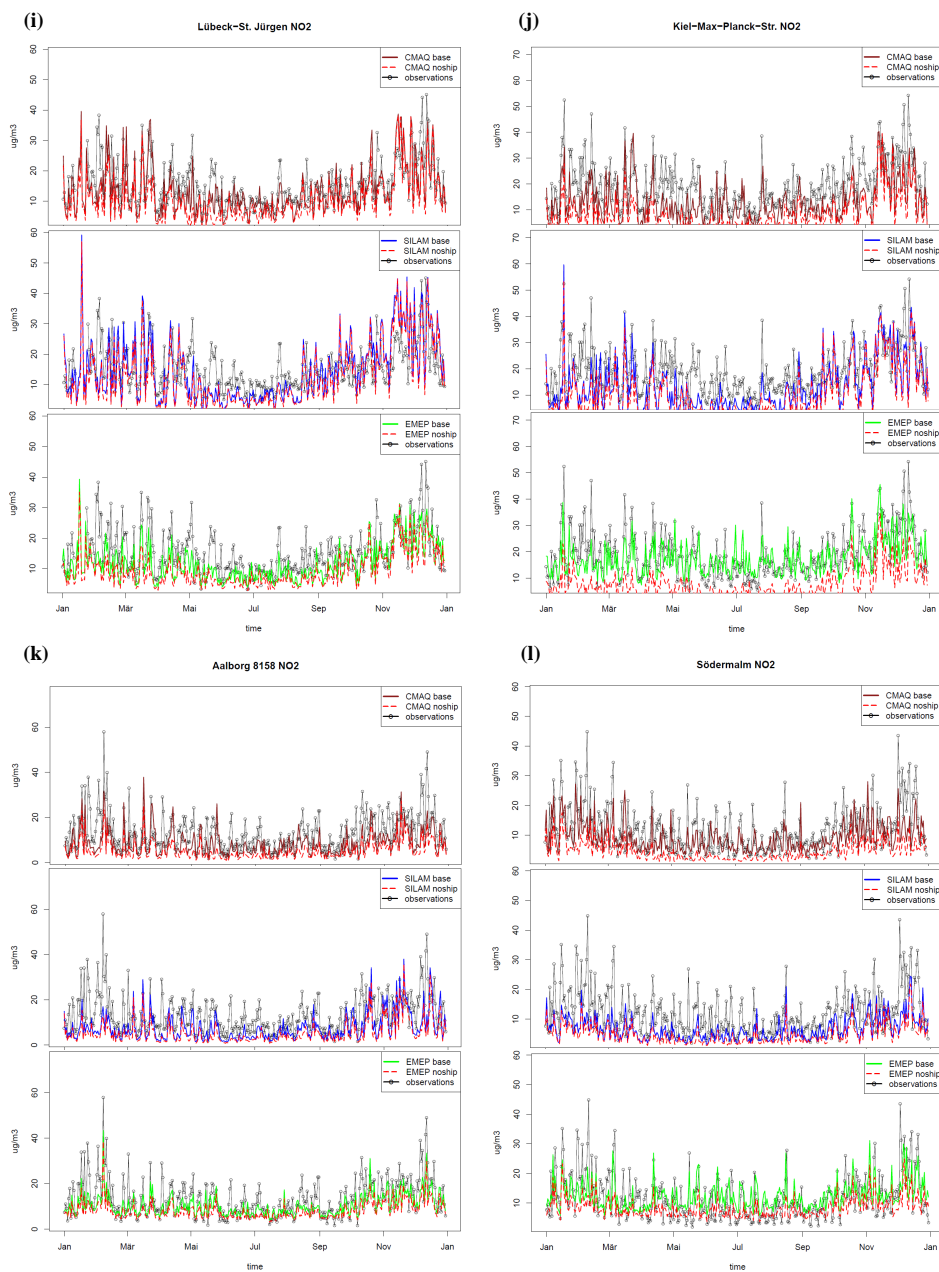


Figure B1. Ship influence on daily mean NO_2 concentration ($\mu\text{g m}^{-3}$): (a) EE0011A; (b) FI00349; (c) SE0066A; (d) DESH023; (e) FI00351; (f) EE0009R; (g) PL0053A; (h) PL0171A; (i) DEMV021; (j) DESH033; (k) DK0053A; (l) SE0022A. Model data from CMAQ (dark red line), SILAM (blue line), EMEP/MSC-W (green line), the respective “noship” run (dashed red line) and observations (black line with open circles).

Table 1. Description of the model setup of the three CTM systems.

Model parameter	CMAQ	SILAM	EMEP/MSC-W
Horizontal grid resolution of the inner nest	4 × 4 km ²	4 × 4 km ²	0.1° × 0.1°
Nesting	D1: 64 km D2: 16 km D3: 4 km	D1: ~70 km D2: ~18 km D3: 4 km	D1: 0.1°
Meteorological driver	COSMO-CLM	WRF	IFS-ECMWF, Cy40r1
Chemical boundary and initial conditions	FMI APTA global reanalysis	FMI APTA global reanalysis	Climatology for ozone
Land-based emissions	SMOKE-EU	SMOKE-EU	ECLIPSE
Ship emissions	STEAM 2 × 2 km ²	STEAM 2 × 2 km ²	STEAM 0.1° × 0.1°
Ship emission time variability	hourly	hourly	monthly

~~Table 2. Vertical profiles of ship emissions in the first seven vertical layers of the three CTM systems. The vertical distribution is given as average fractions of the total emission in each vertical model layer. Heights correspond to the heights of vertical layers in the models for a standard atmosphere.~~

Layer nr.	CMAQ		SILAM		EMEP/MSC-W	
	Height	Fraction	Height	Fraction	Height	Fraction
1	0–42 m	0.24	0–20 m	0.15	0–92 m	1.00
2	42–84 m	0.76	20–50 m	0.13	92–184 m	0
3	84–127 m	0	50–100 m	0.04	184–324 m	0
4	127–180 m	0	100–200 m	0.08	324–525 m	0
5	180–255 m	0	200–300 m	0.08	525–787 m	0
6	255–340 m	0	300–500 m	0.15	787–1116 m	0
7	340–430 m	0	500–1000 m	0.38	1116– 1520 m	0

Table 3. Statistical evaluation of daily mean PM_{2.5} for the year 2012 with all three CTMs.

Station code	N	μ_{Obs}	CMAQ					SILAM					EMEP/MSC-W				
			μ_{Mod}	R	NMB	RMSE	FAC2	μ_{Mod}	R	NMB	RMSE	FAC2	μ_{Mod}	R	NMB	RMSE	FAC2
DEHH008	363	12.6	9.8	0.73	-0.23	7.3	0.73	11.1	0.68	-0.12	7.2	0.86	10.8	0.71	-0.14	7.5	0.87
DEHH059	350	13.2	9.4	0.75	-0.28	7.3	0.65	11.1	0.76	-0.16	6.5	0.86	10.7	0.75	-0.18	7.1	0.87
DEMV019	364	12.9	7.1	0.79	-0.45	8.1	0.42	9.7	0.69	-0.25	7.9	0.77	8.0	0.73	-0.38	7.9	0.59
DEMV021	359	11.2	7.3	0.81	-0.35	6.5	0.55	8.8	0.75	-0.22	6.8	0.80	8.2	0.77	-0.27	6.4	0.73
DESH023	345	11.9	8.2	0.75	-0.31	7.2	0.63	10.0	0.73	-0.16	7.0	0.80	9.1	0.73	-0.23	7.2	0.76
FI00208	347	7.1	9.8	0.53	0.38	7.9	0.59	7.7	0.55	0.09	5.3	0.76	4.2	0.62	-0.41	4.9	0.56
FI00349	355	4.7	3.9	0.62	-0.17	3.1	0.58	5.3	0.55	0.12	4.7	0.61	3.4	0.63	-0.28	3.4	0.73
FI00351	364	7.4	5.2	0.44	-0.30	5.4	0.45	6.3	0.51	-0.14	5.6	0.67	3.4	0.63	-0.54	5.8	0.43
FI00586	357	5.9	4.0	0.64	-0.32	3.6	0.54	6.2	0.49	0.05	5.2	0.57	3.1	0.70	-0.49	4.3	0.55
FI00761	363	6.3	6.3	0.62	0.00	4.5	0.63	6.5	0.44	0.02	5.0	0.70	3.8	0.57	-0.39	4.3	0.63
FI00781	363	6.6	9.5	0.49	0.45	7.1	0.62	10.7	0.33	0.62	8.4	0.48	5.0	0.63	-0.24	4.0	0.82
LT00044	357	9.8	12.0	0.59	0.22	9.0	0.65	12.1	0.73	0.23	7.2	0.75	8.9	0.69	-0.09	6.4	0.78
SE0001A	338	11.0	6.4	0.61	-0.42	7.0	0.49	7.9	0.64	-0.28	6.6	0.67	6.9	0.71	-0.37	6.4	0.64
SE0011R	340	6.9	5.7	0.51	-0.17	4.8	0.57	7.0	0.65	0.01	5.1	0.75	5.8	0.68	-0.16	4.8	0.73
SE0012R	364	5.4	4.1	0.50	-0.24	3.8	0.52	5.4	0.58	-0.01	3.7	0.74	3.5	0.67	-0.36	3.3	0.64
SE0022A	358	4.8	6.7	0.41	0.39	5.4	0.67	6.9	0.48	0.42	4.9	0.66	5.0	0.66	0.04	3.1	0.86
SE0066A	345	3.3	4.1	0.50	0.25	3.6	0.58	5.3	0.44	0.60	4.5	0.47	3.2	0.54	-0.04	2.4	0.66

Table 2. Spatial averages of the annual mean concentrations of NO₂, O₃, SO₂, PM_{2.5} and EC in $\mu\text{g m}^{-3}$ for the study domain (Baltic Sea region as in Fig. 1).

CTM	NO ₂	O ₃	SO ₂	PM _{2.5}	EC
CMAQ	3.49	54.1	1.11	4.84	0.16
SILAM	4.42 <u>4.82</u>	57.6 <u>56.7</u>	1.61 <u>1.65</u>	5.95 <u>5.94</u>	0.15
EMEP/MSC-W	3.00	66.6	0.56	4.01	0.13

Table 3. Relative ship contribution to the spatial average of annual mean NO₂, O₃, SO₂, PM_{2.5} and EC in percent for the study domain (Baltic Sea region as in Fig. 1). Values in brackets denote the average relative ship contribution in the coastal land areas of the domain.

CTM	NO ₂	O ₃	SO ₂	PM _{2.5}	EC
CMAQ	28.3	-0.4	14.5	6.5	7.3
	<u>(20.3)</u>	<u>(0.4)</u>	<u>(10.1)</u>	<u>(5.7)</u>	<u>(5.0)</u>
SILAM	21.9 <u>25.6</u>	1.2 <u>-0.4</u>	10.0 <u>11.8</u>	4.3 <u>4.1</u>	5.6 <u>5.4</u>
	<u>(17.6)</u>	<u>(0.1)</u>	<u>(8.7)</u>	<u>(3.1)</u>	<u>(4.1)</u>
EMEP/MSC-W	21.8	-0.1	19.1	5.7	5.3
	<u>(16.1)</u>	<u>(0.4)</u>	<u>(14.1)</u>	<u>(4.6)</u>	<u>(3.6)</u>

Table 4. Spatial averages of seasonal mean concentrations of EC ($\mu\text{g m}^{-3}$) from the “base” simulation and ship contributions to EC levels ($\mu\text{g m}^{-3}$) for the study domain (Baltic Sea region as in Fig. 1). Mean values are given for spring (March to May; MAM), summer (JJA), autumn (Sept. to Nov.; SON) and winter (January, February and December 2012; DJF). Values in brackets denote the seasonal mean concentrations in the coastal land areas of the domain.

Contribution	CTM	MAM	JJA	SON	DJF
All emissions	CMAQ	0.134	0.081	0.154	0.277
		<u>(0.134)</u>	<u>(0.077)</u>	<u>(0.158)</u>	<u>(0.291)</u>
	SILAM	0.136 <u>0.137</u>	0.109 <u>0.112</u>	0.183 <u>0.184</u>	0.166 <u>0.165</u>
		<u>(0.102)</u>	<u>(0.081)</u>	<u>(0.139)</u>	<u>(0.134)</u>
	EMEP/MSC-W	0.111	0.072	0.150	0.191
	<u>(0.102)</u>	<u>(0.065)</u>	<u>(0.146)</u>	<u>(0.196)</u>	
Ship emissions	CMAQ	0.013	0.014	0.009	0.008
		<u>(0.008)</u>	<u>(0.009)</u>	<u>(0.007)</u>	<u>(0.007)</u>
	SILAM	0.007 <u>0.008</u>	0.011 <u>0.013</u>	0.007 <u>0.009</u>	<u>0.002</u>
		<u>(0.003)</u>	<u>(0.005)</u>	<u>(0.003)</u>	<u>(0.003)</u>
	EMEP/MSC-W	0.008	0.009	0.005	0.004
	<u>(0.004)</u>	<u>(0.005)</u>	<u>(0.003)</u>	<u>(0.003)</u>	