Reply to Editor Comment

We thank Dr Huan Liu for positive evaluation of the manuscript. The Editor Comment has been submitted twice (EC1 and RC2), here we reply to RC2. We have addressed the technical questions brought up by the Reviewer below, with specific pointers to changed parts of the manuscript.

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.***

Reply: 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 (Robertson et al., 2007). We note that multi-generational aging chemistry for the semi-volatile POA was introduced in CMAQ v5.2, derived from 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)."

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

Reply: 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.

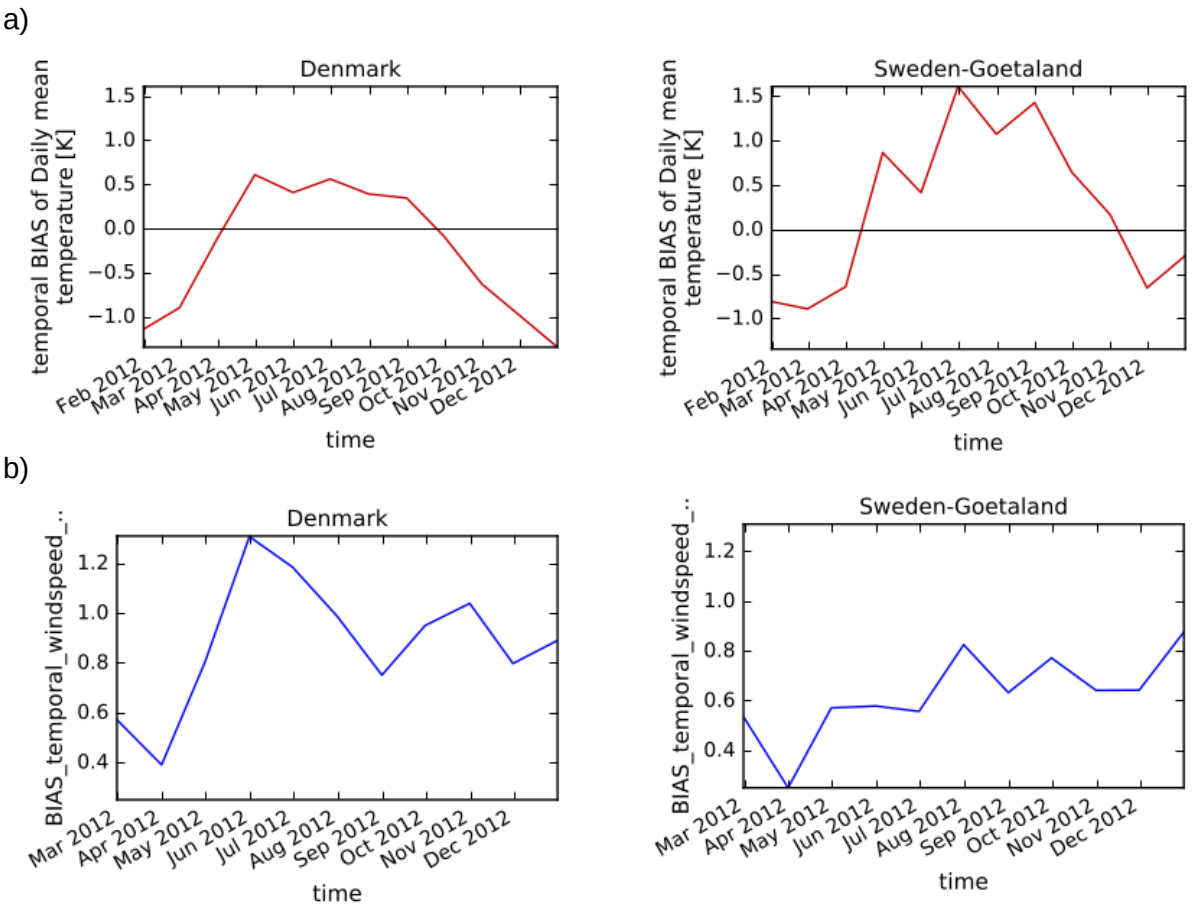

Figure C1: Evaluation of COSMO-CLM data: temporal bias a) of the monthly mean 2 m air temperature in Denmark (left) and southern Sweden (right); and b) of the monthly mean 10 m wind speed in Denmark (left) and southern Sweden (right).

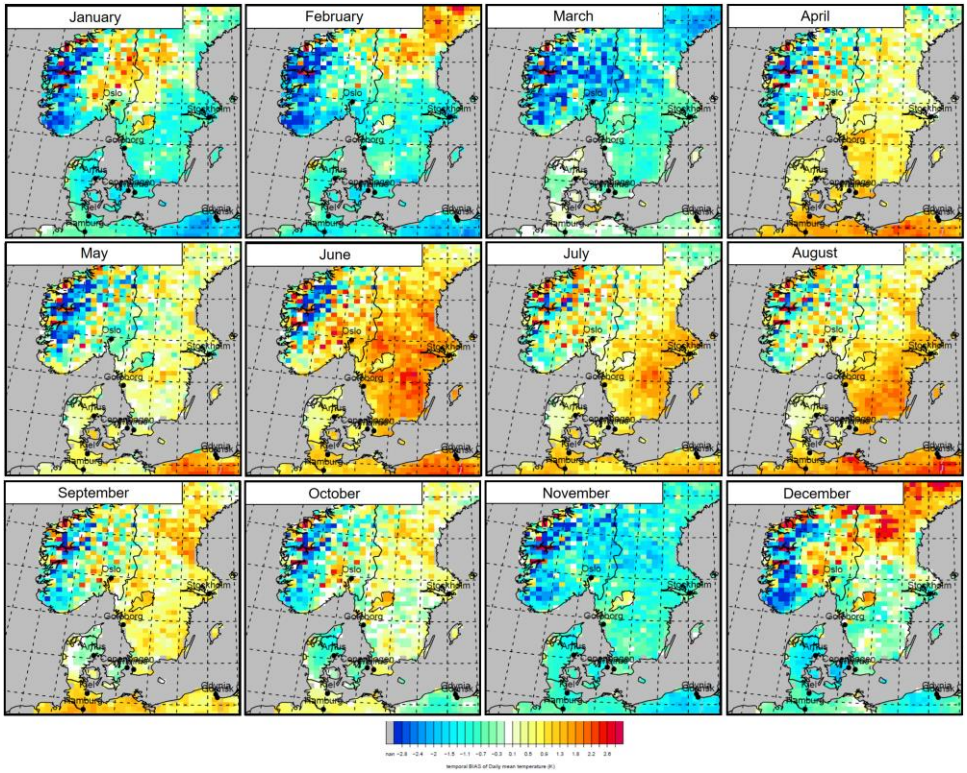

Figure C2: Evaluation of COSMO-CLM data: spatial distribution of the temporal bias of the monthly mean 2 m air temperature in the southern part of the Baltic Sea region.

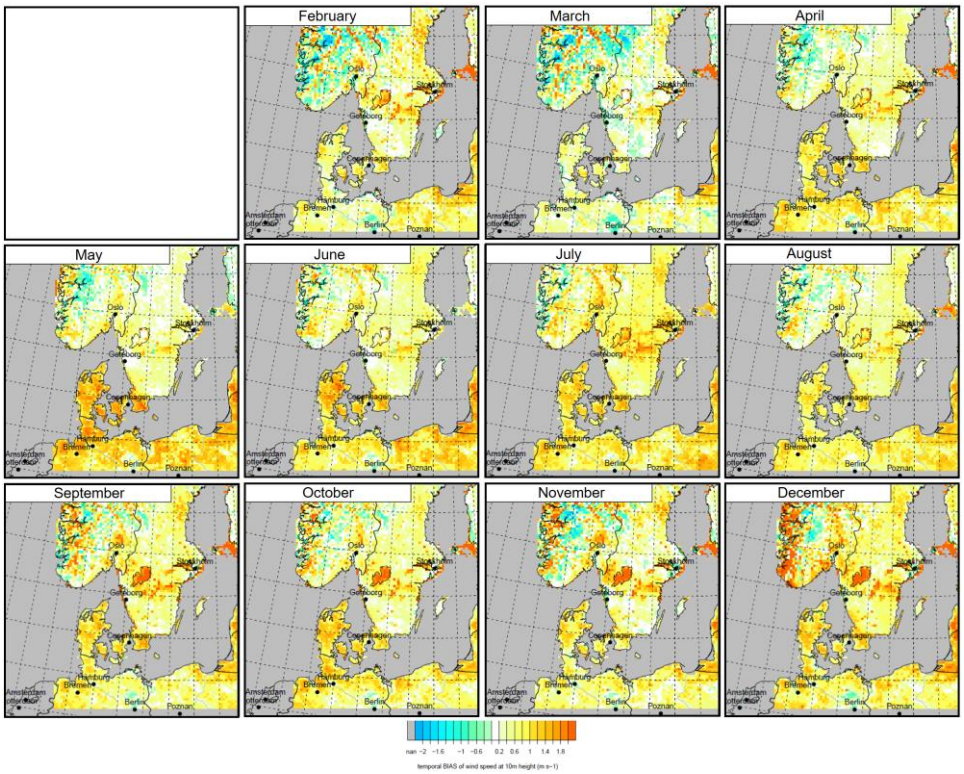

Figure C3: Evaluation of COSMO-CLM data: spatial distribution of the temporal bias of the monthly mean 10 m wind speed in the southern part of the Baltic Sea region. Missing observation data for January 2012.

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.***

Response:

Reply: 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.***

Reply: 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 "NOx-limited regime in the model".***

Reply: 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.

References:

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