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Supplement of

Impact of a nitrogen emission control area (NECA) on the future air quality and nitrogen deposition to seawater in the Baltic Sea region

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S1. Methodology and results for the evaluation of modelled precipitation

The precipitation amount and frequency from COSMO-CLM output was compared against daily precipitation measurements from rain gauge stations operated by the Swedish Meteorological and Hydrological Institute (SMHI). 1804 precipitation stations in Sweden were recording daily precipitation sums during 2012; each measurement starting at 6 a.m. UTC on the previous day until 6 a.m. UTC of the current day and is available from the SMHI opendata portal (<http://opendata-catalog.smhi.se/explore/>). The station network for daily precipitation collection densely covers the regions of south and middle Sweden (Götaland and Svealand) where each station represents an area of about $10 \times 10 \text{ km}^2$, with lower density of stations in the northern part of Sweden (Södra Norrland and Norra Norrland). Precipitation measurements are known to suffer from evaporation and wind loss, especially on days with very low rain rates (Nespor and Sevruk, 1998). Therefore, a precipitation threshold of 1 mm d^{-1} is commonly used for the definition of dry days (WMO, 2011). In the present comparison of modelled with observed precipitation no observational threshold was applied in the data analysis. However, in order to determine the performance of COSMO-CLM with respect to the prediction of the number of days without rainfall and hence no wet deposition is examined based on the definition of dry days by a threshold of 0.1 mm d^{-1} for the model and observational data.

Note for the comparison of modelled and observed precipitation, that the model data is a grid average (either of grid boxes with 0.11 degree or 0.025 degree cell width) whereas the rain gauge data represent a point measurement. Hence, the high-resolution output of COSMO-CLM should better capture the variability of the precipitation measurements. The model-observation comparison was done for the three different configurations of COSMO-CLM: 0.11 degree grid resolution with Tiedtke scheme for convection (“011”), 0.025 degree grid resolution with Tiedtke scheme for convection (“0025_Tiedtke”), and 0.025 degree grid resolution with convection-permitting configuration (“0025_convper”). Fig. S1 shows the

monthly precipitation amounts from the three model configurations of the summer months for the Baltic Sea and North Sea region and compares to the measured precipitation amount at the Swedish rain gauge stations (circles filled with colour indicating the observed value).

Fig. S2 shows the probability distributions of the differences in seasonal averaged (winter months and summer months of 2012) daily precipitation sums from the three model outputs and the observation data in the four regions of Sweden. The percentage fraction of days with zero difference between model and observation and days with difference of 0.1 mm d^{-1} (“delta01” days), corresponding to the threshold value, was calculated. Large deviations between model and observations ($> 10 \text{ mm d}^{-1}$) are rare for all three configurations. For “0025_convper”, the percentage fraction of days with model-observation deviations below the threshold (“delta01” days) is in the range of 18–34 % in winter and 29–38 % in summer, depending on the region.

The fraction of observed dry days (daily sum $< 0.1 \text{ mm d}^{-1}$), as average of all stations of one region, of summer and winter (Table S1) is always higher than the fraction of “delta01” days in the corresponding model data. This implies that COSMO-CLM, on a statistical average, predicts more precipitation days than observed. For “0025_convper”, the number of precipitation days, depending on the region, is 9–18 % higher in summer and 9–11 % higher in winter than observed (assuming that the “delta01” days correspond to dry days).

By summing up the model-observation differences of all days in summer and winter averaged over all stations of one region, the model bias for precipitation amounts was determined (Table S2). Note that in this study winter is defined as JFD, including the months January, February and December from 2012 since the simulation was only done for one year. In summer, precipitation in “0025_convper” has a relative bias of -25 %, -19 %, 51 %, and 42 % in Götaland, Svealand, S. Norland, N. Norland, respectively, compared to observed

precipitation amounts. In winter, precipitation from “0025_convper” has a much lower relative bias (-4 %, -7 %, 3 %, and 32 % in Götaland, Svealand, S. Norland, N. Norland, respectively).

The probability distribution of differences for all months for Götaland is shown in Fig. S3.

The convection-permitting configuration “0025_convper” reduces the frequency of negative differences in the range of 0.1–4 mm d⁻¹ for daily rainfall in June compared to

“0025_Tiedtke”. The frequencies of negative differences between observation and model is similar for all summer months, while observed total monthly precipitation amount is highest in June (Götaland station average, June: 111 mm, July: 87 mm, August: 68 mm). COSMO-CLM performs better in the cold season (from October to March) in Götaland when differences between observations and model are in the range between -4 and +6 mm d⁻¹ for more than 90 % of the time (Fig. S3).

S2. Methodology and results for the evaluation of modelled wet deposition of nitrogen

The modelled wet deposition of nitrogen from CMAQ output was compared against data from the EMEP monitoring programme (Tørseth et al., 2012; EMEP, 2014). Observation data was obtained from the EBAS database (<http://ebas.nilu.no/>). Modelled wet deposition of nitrogen was separately compared for nitrate, NO_3^- (WNO_3), representing oxidised nitrogen and modelled wet deposition of ammonium, NH_4^+ (WNH_4), representing reduced nitrogen. Modelled wet deposition of HNO_3 was included in WNO_3 and modelled wet deposition of NH_3 was included in WNH_4 , because it is assumed that the gases are partially or fully dissolved in the sampled rainwater. The summation also gives a more robust estimate of the wet deposition of oxidised and reduced nitrogen. Measured concentrations of NO_3^- and NH_4^+ in rainwater were converted into nitrogen deposition per area by the amount of rainwater measured at the respective station. Daily sums of wet deposition were calculated from the volume-weighted concentrations multiplied by the precipitation amounts. The comparison of the daily sum of wet deposition was done in terms of mean values (μ_{Mod} and μ_{Obs}), the Spearman's correlation coefficient (R_{Spr}) and the normalized mean bias (NMB). Only days with predicted and observed rain events in common were included in the comparison. Several stations in the Baltic Sea region had only few measurements during the period. Stations with less than seven model-observation pairs were excluded from the statistical analysis. CMAQ model data from the intermediate grid (CD16) and from the high-resolution grid (CD04) were evaluated separately.

Plots in Fig. 3b-g show the time series modelled and observed daily sums of WNO_3 at selected stations, while all other stations are shown in Fig. S4. WNO_3 is underestimated at all stations included in the statistical analysis (Table S3), most severely at the Finnish stations and at Zingst (NMB between -0.75 and -0.90).

WNH₄ is underestimated at all stations included in the statistical analysis (Table S4; corresponding time series are plotted in Fig. S5). The underestimation is highest for Zingst and the Finnish stations, as for WNO₃.

To account for the fact that the days with predicted rain often do not correspond to days with observed rain, seasonal averages (spring, summer and autumn) were calculated for WNO₃ (Table S5) and WNH₄ (Table S6) independently for CD04 model data and observation data. The joint underestimation of WNO₃ and WNH₄ at Zingst and the Finnish stations is confirmed in this analysis. At Zingst, seasonal averages of WNO₃ based on model data are lower by -52 % to -63 % and seasonal averages of WNH₄ based on model data are lower by -64 % to -79 % than the corresponding seasonal averages based on observations. The ratio of modelled to observed seasonal average of WNO₃ shows little variation for the three seasons (0.37–0.48). At Preila, a coastal station in Lithuania, both observed averages of WNO₃ and WNH₄ are underestimated in spring and autumn, but not in summer. The ratio of modelled to observed seasonal averages of WNO₃ and WNH₄ show a consistent seasonal pattern at the nine stations (Fig. S6), pointing to the formation and atmospheric transport of particulate ammonium nitrate as common cause.

S3. Methodology and results for the evaluation of modelled air pollutant concentrations

The modelled surface air concentrations of O₃, NO₂, SO₂ and PM_{2.5} from CMAQ output of the 4-km resolution grid were compared against measurements at regional background stations of the EMEP monitoring programme available from the EBAS database. In addition to the statistical indicators used in the evaluation of deposition, the root mean square error of the modelled values (RMSE) was included in the evaluation of air concentrations, a frequently used measure of the differences between values predicted by a model and the values actually observed. The evaluation was done for the entire year 2012 and separately for summer (JJA) 2012. In the context of this evaluation of predicted air pollutant concentrations, a correlation coefficient of more than 0.5 was considered to indicate a correlation between modelled and observed time series, while values of 0.7 and above was considered as a good correlation.

The results for the statistical evaluation of modelled daily mean O₃ concentrations are summarized in Table S7. Modelled daily means of O₃ are in good agreement with measurements at all stations ($R_{Spr} = 0.75$, RMSE = 6.8 ppbv, both as average of all stations; NMB range: -0.16 to -0.02) when the entire year is considered. In summer, ozone is slightly underestimated at the stations in the southern part of the Baltic Sea region (NMB range: -0.23 to -0.12). The overall agreement in summer is, however, fairly good ($R_{Spr} = 0.62$, RMSE = 6.9 ppbv; each as average of all stations).

The evaluation of modelled NO₂ based on daily concentrations for the entire year and for summer (Table S8) indicates a better performance of CMAQ over the entire year than over summer alone. A good correlation is obtained at 9 out of 12 stations for the entire year, whereas only 5 stations show a good correlation in summer. MNB is positive at 11 stations for the entire year and at 8 stations for summer. The CD04 simulations predict slightly higher NO₂ concentrations than observed at most stations (NMB range: -0.28–0.44 for the year;

NMB range: -0.31–0.83 for JJA; average of all stations). RMSE is in the range 1.0–3.2 ppbv for the year and in the range 0.2–3.5 ppbv for summer.

Simulated SO₂ daily mean concentrations are correlated with the observed daily mean concentrations at all stations of the Baltic Sea region for the entire year but in summer they are not correlated at several stations (Table S9). The associated RMSE is relatively high (RMSE = 1.01 ppbv for the year, RMSE = 0.41 ppbv for JJA; average of all stations) but the summer RMSE is lower than in the multi-model study by Giordano et al. (2015) (RMSE range: 2.17–2.34 ppbv). Observed SO₂ concentrations are generally overestimated (NMB range: -0.04–1.62 for the year; NMB range: -0.07–1.84 for JJA). In particular, summer mean SO₂ at the remote stations Ähtäri II (Finland) and Preila (Lithuania) is overestimated by a factor of 2–3.

Data from eight stations was available for the comparison of modelled against observed PM_{2.5} (Table S10). For the entire year CMAQ performs quite well in the prediction of daily mean PM_{2.5} ($R_{Spr} = 0.57$, NMB = -0.22, RMSE = 5.6 $\mu\text{g m}^{-3}$; each as average of all stations). In the summer period, PM_{2.5} is underestimated at all stations (average NMB = -0.60).

Table S1: Percentage fraction of dry days, as observed and as predicted by COSMO-CLM model with configuration “0025_convper” for summer (JJA) and for winter (JFD) of 2012 in the four regions of Sweden. Values are given as average of all stations in a region. For the fraction of predicted dry days it is assumed that the “delta01” days (days with model-observation difference below the threshold of 0.1 mm d⁻¹) correspond to dry days.

Region	Summer		Winter	
	Predicted dry days [%]	Observed dry days [%]	Predicted dry days [%]	Observed dry days [%]
Göteborg	38	47	34	43
Svealand	29	41	32	42
S. Norrland	31	47	23	34
N. Norrland	31	49	18	28

Table S2: Precipitation bias, calculated as difference between model and observation (M-O; in mm) for summer (JJA) and for winter (JFD) of 2012. Values are given as average of all stations in a region.

Region	Precipitation bias (M-O) [mm] in summer			Precipitation bias (M-O) [mm] in winter		
	011	0025_ Tiedtke	0025_ convper	011	0025_ Tiedtke	0025_ convper
Göteborg	-99	-35	-67	-1	0	-8
Svealand	-83	-40	-61	20	-4	-11
S. Norrland	10	111	132	50	18	5
N. Norrland	49	89	101	43	34	53

Table S3: Statistical comparison of daily sums of WNO₃ for stations of the EMEP monitoring network in the Baltic Sea region. CMAQ model results for the CD16 and CD04 grid domains are evaluated separately. Statistical indicators include mean values of model (μ_{Mod}), mean values of observations (μ_{Obs}), Spearman's correlation coefficient (R_{Spr}), and normalized mean bias (NMB). Unit of mean values is mg(N) m⁻² d⁻¹. N is the number of samples where precipitation occurred both in the simulation and in the observation. Only stations with more than 7 samples were considered (for station Hailuoto II it is less than 7 for the CD04 grid).

Station, Code	CD16					CD04				
	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB
Zingst, DE0009R	14	-0.09	0.65	5.23	-0.88	9	0.08	1.21	6.03	-0.80
Råö SE0014R	86	0.49	2.37	3.21	-0.26	77	0.55	1.76	2.95	-0.40
Leba, PL0004R	72	0.26	1.56	3.41	-0.54	58	0.40	1.48	3.78	-0.61
Diabla Gora, PL0005R	75	0.29	1.25	2.42	-0.48	63	0.01	1.61	3.02	-0.47
Ähtäri, FI0004R	12	0.06	0.75	3.57	-0.79	11	0.81	0.78	3.16	-0.75
Virolahti II FI0017R	17	-0.01	0.92	5.93	-0.85	14	-0.17	1.07	5.42	-0.80
Hailuoto II, FI0053R	13	0.32	0.69	3.07	-0.78	5	–	–	–	–
Lahemaa, EE0009R	70	0.31	1.05	1.66	-0.37	50	0.27	1.10	1.84	-0.40
Preila LT0015R	58	0.32	1.46	2.66	-0.45	44	0.40	2.04	2.733	-0.25

Table S4: Statistical comparison of daily sums of WNH_4 for stations of the EMEP monitoring network in the Baltic Sea region. CMAQ model results for the CD16 and CD04 grid domains are evaluated separately. Statistical indicators include mean values of model (μ_{Mod}), mean values of observations (μ_{Obs}), Spearman's correlation coefficient (R_{Spr}), and normalized mean bias (NMB). Unit of mean values is $\text{mg(N)} \text{ m}^{-2} \text{ d}^{-1}$. N is the number of samples where precipitation occurred both in the simulation and in the observation. Only stations with more than 7 samples were considered (for station Hailuoto II it is less than 7 for the CD04 grid).

Station, Code	CD16					CD04				
	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB
Zingst, DE0009R	12	0.23	0.76	8.88	-0.91	8	0.52	1.54	9.60	-0.84
Råö SE0014R	81	0.52	2.03	3.28	-0.38	63	0.38	1.83	3.42	-0.46
Leba, PL0004R	69	0.35	1.31	4.21	-0.69	57	0.24	1.29	4.64	-0.72
Diabla Gora, PL0005R	68	0.20	1.28	3.39	-0.62	56	0.06	1.86	3.77	-0.51
Ähtäri, FI0004R	12	0.07	0.68	2.59	-0.74	9	0.58	0.81	2.82	-0.71
Virolahti II FI0017R	18	-0.07	0.70	4.95	-0.86	13	-0.03	1.02	4.68	-0.78
Hailuoto II, FI0053R	13	-0.18	0.60	3.50	-0.83	4	–	–	–	–
Lahemaa, EE0009R	47	0.46	1.09	1.43	-0.24	36	0.29	1.12	1.76	-0.37
Preila LT0015R	53	0.48	1.23	2.26	-0.45	40	0.45	2.04	2.53	-0.19

Table S5: Seasonal averages of wet deposition of nitrate for stations of the EMEP monitoring network in the Baltic Sea region. CMAQ model results were taken from the CD04 grid domains. For spring (MAM), summer (JJA) and autumn (SON), the mean value of model (μ_{Mod}) and mean value of observations (μ_{Obs}) is given based on daily sums. Unit of mean values is mg(N) m⁻² d⁻¹. Observational and model data was evaluated independently. N_{Obs} is the number of observations in the respective season, used to calculate μ_{Obs} . N_{Mod} is the number of days with simulated precipitation in the respective season, used to calculate μ_{Mod} .

Station, Code	Spring				Summer				Winter			
	N_{Mod}	N_{Obs}	μ_{Mod}	μ_{Obs}	N_{Mod}	N_{Obs}	μ_{Mod}	μ_{Obs}	N_{Mod}	N_{Obs}	μ_{Mod}	μ_{Obs}
Zingst, DE0009R	9	11	1.71	3.54	18	10	2.25	5.71	22	13	1.85	4.98
Råö SE0014R	13	24	3.38	3.48	15	29	1.51	3.80	42	48	2.23	2.83
Leba, PL0004R	14	24	1.76	1.47	10	31	2.88	5.27	33	58	1.43	2.21
Diabla Gora, PL0005R	20	29	1.75	3.64	19	25	2.22	4.06	30	46	1.60	3.06
Ähtäri, FI0004R	7	9	1.09	3.25	14	9	0.92	2.73	17	12	1.36	3.46
Virolahti II FI0017R	13	11	1.98	3.30	14	9	1.33	3.48	36	16	1.26	6.50
Hailuoto II, FI0053R	9	11	0.70	3.16	11	8	0.86	1.61	23	15	1.03	3.24
Lahemaa, EE0009R	17	29	1.37	1.64	17	21	1.24	1.27	24	39	1.32	2.11
Preila LT0015R	14	10	1.52	3.09	17	24	2.63	2.16	29	41	1.61	3.04

Table S6: Seasonal averages of wet deposition of ammonium for stations of the EMEP monitoring network in the Baltic Sea region. CMAQ model results were taken from the CD04 grid domains. For spring (MAM), summer (JJA) and autumn (SON), the mean value of model (μ_{Mod}) and mean value of observations (μ_{Obs}) is given based on daily sums. Unit of mean values is $\text{mg(N) m}^{-2} \text{ d}^{-1}$. Observational and model data was evaluated independently. N_{Obs} is the number of observations in the respective season, used to calculate μ_{Obs} . N_{Mod} is the number of days with simulated precipitation in the respective season, used to calculate μ_{Mod} .

Station, Code	Spring				Summer				Winter			
	N_{Mod}	N_{Obs}	μ_{Mod}	μ_{Obs}	N_{Mod}	N_{Obs}	μ_{Mod}	μ_{Obs}	N_{Mod}	N_{Obs}	μ_{Mod}	μ_{Obs}
Zingst, DE0009R	9	11	2.36	7.73	19	11	2.01	9.68	19	14	2.30	6.33
Råö SE0014R	13	24	3.87	5.03	17	11	1.02	1.64	34	40	2.17	2.42
Leba, PL0004R	13	22	2.12	2.20	14	7	1.08	1.94	28	55	1.33	2.21
Diabla Gora, PL0005R	23	27	2.32	6.18	14	9	1.44	3.51	28	39	1.47	2.44
Ähtäri, FI0004R	7	9	1.11	3.03	14	7	1.08	1.94	17	11	0.98	2.28
Virolahti II FI0017R	14	11	2.61	2.98	14	9	1.44	3.51	28	15	1.12	5.62
Hailuoto II, FI0053R	9	11	0.75	3.86	10	8	1.00	1.51	18	14	0.74	3.64
Lahemaa, EE0009R	17	22	1.64	2.01	17	11	1.02	1.64	23	17	0.90	1.78
Preila LT0015R	15	10	1.60	4.19	16	22	3.24	2.57	23	39	1.25	2.42

Table S7: Statistical evaluation of modelled O₃ concentrations (in ppbv) with measurements of the EMEP monitoring network in the Baltic Sea region based on daily means for the entire year and for summer (JJA). Statistical indicators: mean values of model (μ_{Mod}), mean values of observations (μ_{Obs}), Spearman's correlation coefficient (R_{Spr}), normalized mean bias (NMB) and root mean square error of the modelled values (RMSE; in ppbv). All stations of the EMEP network located within the CD04 grid domain with available measurements were considered. N is the number of the available daily mean measurements at the respective station.

Station Code	CD04 annual						CD04 summer					
	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	RMSE	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	RMSE
Zingst, DE0009R	366	0.80	27.3	27.8	-0.02	5.9	92	0.64	29.0	33.0	-0.12	6.5
Keldsnor, DK0005R	357	0.66	26.3	28.1	-0.07	7.2	86	0.29	26.8	32.9	-0.18	9.7
Risoe, DK0012R	366	0.74	26.1	29.3	-0.11	6.1	92	0.48	26.9	35.0	-0.23	10.0
Ähtäri II, FI0037R	358	0.79	23.8	24.8	-0.04	5.4	92	0.78	23.6	22.3	0.06	4.4
Virolahti II, FI0017R	358	0.70	25.0	26.6	-0.06	7.0	91	0.77	26.1	24.5	0.07	5.5
Utö, FI0009R	362	0.74	28.1	31.1	-0.09	6.4	90	0.68	28.9	34.6	-0.17	7.6
Rucava, LV0010R	347	0.73	27.8	33.1	-0.16	9.5	92	0.67	30.5	32.3	-0.06	6.7
Vilsandi, EE0011R	361	0.80	28.8	31.7	-0.09	6.1	90	0.83	31.1	35.7	-0.13	6.3
Lahemaa, EE0009R	365	0.69	25.4	26.5	-0.04	6.7	91	0.69	26.9	26.4	0.02	5.3
Preila, LT0015R	364	0.77	29.2	29.8	-0.02	6.7	91	0.48	33.2	33.5	-0.01	7.2

Table S8: Statistical evaluation of modelled NO₂ concentrations (in ppbv) with measurements of the EMEP monitoring network in the Baltic Sea region based on daily means for the entire year and for summer (JJA). Statistical indicators: mean values of model (μ_{Mod}), mean values of observations (μ_{Obs}), Spearman's correlation coefficient (R_{Spr}), normalized mean bias (NMB) and root mean square error of the modelled values (RMSE; in ppbv). All stations of the EMEP network located within the CD04 grid domain with available measurements were considered. N is the number of the available daily mean measurements at the respective station.

Station Code	CD04 annual						CD04 summer					
	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	RMSE	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	RMSE
Zingst, DE0009R	355	0.73	3.8	3.4	0.10	2.04	88	0.58	2.9	2.4	0.22	1.50
Keldsnor, DK0005R	331	0.84	5.1	4.0	0.27	3.24	86	0.82	4.9	3.2	0.52	3.46
Anholt, DK0008R	283	0.81	3.7	2.6	0.42	2.67	92	0.80	3.6	2.0	0.83	2.91
Risoe, DK0012R	351	0.83	4.7	4.6	0.02	1.97	88	0.83	3.5	2.9	0.19	1.19
Ähtäri II, FI0037R	349	0.86	1.0	1.2	-0.22	1.05	78	0.47	0.3	0.4	-0.31	0.18
Virolahti II, FI0017R	365	0.65	2.1	2.6	-0.18	2.60	92	0.54	1.5	1.5	-0.01	0.90
Hyytiälä, FI0050R	142	0.90	1.3	1.8	-0.28	1.63	32	0.70	0.3	0.4	-0.10	0.19
Utö, FI0009R	331	0.71	1.9	1.6	0.18	1.26	87	0.66	2.1	1.4	0.48	1.50
Rucava, LV0010R	357	0.74	1.9	1.3	0.44	1.35	83	0.30	1.0	0.7	0.36	0.49
Vilsandi, EE0011R	340	0.69	1.6	1.3	0.24	1.12	89	0.49	1.4	0.8	0.71	1.44
Lahemaa, EE0009R	335	0.76	1.8	1.5	0.20	1.21	84	0.71	1.2	0.8	0.59	0.75
Preila, LT0015R	333	0.65	1.8	1.8	0.04	1.54	89	0.42	1.0	1.2	-0.18	0.59

Table S9: Statistical evaluation of modelled SO₂ concentrations (in ppbv) with measurements of the EMEP monitoring network in the Baltic Sea region based on daily means for the entire year and for summer (JJA). Statistical indicators: include mean values of model (μ_{Mod}), mean values of observations (μ_{Obs}), Spearman's correlation coefficient (R_{Spr}), normalized mean bias (NMB) and root mean square error of the modelled values (RMSE; in ppbv). All stations of the EMEP network located within the CD04 grid domain with available measurements were considered. N is the number of the available daily mean measurements at the respective station.

Station Code	CD04 annual						CD04 summer					
	N	R_{Spr}	μ_{Mo}	μ_{Obs}	NMB	RM	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	RMS
Zingst, DE0009R	366	0.54	1.21	0.87	0.40	0.98	92	0.54	0.75	0.80	-0.07	0.37
Anholt, DK0008R	351	0.73	0.87	0.37	1.33	0.79	92	0.73	0.76	0.43	0.76	0.62
Risoe, DK0012R	350	0.70	1.03	0.42	1.46	0.91	92	0.73	0.68	0.37	0.81	0.45
Ähtäri II, FI0037R	50	0.61	0.50	0.35	1.15	0.40	12	0.04	0.19	0.07	1.84	0.17
Virolahti II, FI0017R	282	0.72	1.24	1.26	-0.02	1.22	50	0.47	0.74	0.43	0.73	0.55
Utö, FI0009R	318	0.74	0.78	0.65	0.20	0.57	75	0.60	0.59	0.50	0.18	0.41
Rucava, LV0010R	359	0.58	1.25	0.48	1.62	1.50	92	0.29	0.60	0.35	0.71	0.52
Vilsandi, EE0011R	283	0.69	0.84	0.58	0.44	0.78	67	0.39	0.48	0.39	0.24	0.25
Lahemaa, EE0009R	270	0.57	1.13	1.17	-0.04	1.45	75	0.42	0.61	0.58	0.05	0.41
Preila, LT0015R	302	0.57	1.29	0.60	1.15	1.47	92	0.30	0.55	0.24	1.28	0.43

Table S10: Statistical evaluation of modelled PM_{2.5} concentrations (in $\mu\text{g m}^{-3}$) with measurements of the EMEP monitoring network in the Baltic Sea region based on daily means for the entire year and for summer (JJA). Statistical indicators: include mean values of model (μ_{Mod}), mean values of observations (μ_{Obs}), Spearman's correlation coefficient (R_{Spr}), normalized mean bias (NMB) and root mean square error of the modelled values (RMSE; in $\mu\text{g m}^{-3}$). All stations of the EMEP network located within the CD04 grid domain with available measurements were considered. N is the number of the available daily mean measurements at the respective station.

Station Code	CD04 annual						CD04 summer					
	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	RMSE	N	R_{Spr}	μ_{Mod}	μ_{Obs}	NMB	RMSE
Råö SE0014R	351	0.49	4.9	5.6	-0.13	4.4	87	0.36	2.5	4.9	-0.50	3.5
Diabla Gora, PL0005R	365	0.82	7.8	13.8	-0.44	8.5	92	0.70	2.5	8.7	-0.71	6.8
Vavihill, SE0011R	252	0.55	6.3	8.2	-0.23	5.2	84	0.62	2.8	6.5	-0.58	4.5
Aspvreten, SE0012R	254	0.51	4.9	6.5	-0.24	4.3	78	0.61	1.9	6.0	-0.68	4.7
Utö, FI0009R	357	0.63	3.9	5.2	-0.26	3.4	90	0.71	1.8	5.2	-0.65	3.8
Rucava, LV0010R	327	0.29	6.8	10.8	-0.37	10.4	78	-0.19	2.5	7.5	-0.66	6.5
Vilsandi, EE0011R	342	0.67	4.6	5.5	-0.16	3.9	84	0.73	2.0	4.1	-0.52	3.0
Lahemaa, EE0009R	343	0.60	6.0	5.5	0.09	4.6	87	0.32	1.8	3.6	-0.50	2.7

COSMO-CLM and Observations: Monthly Summer Precipitation 2012

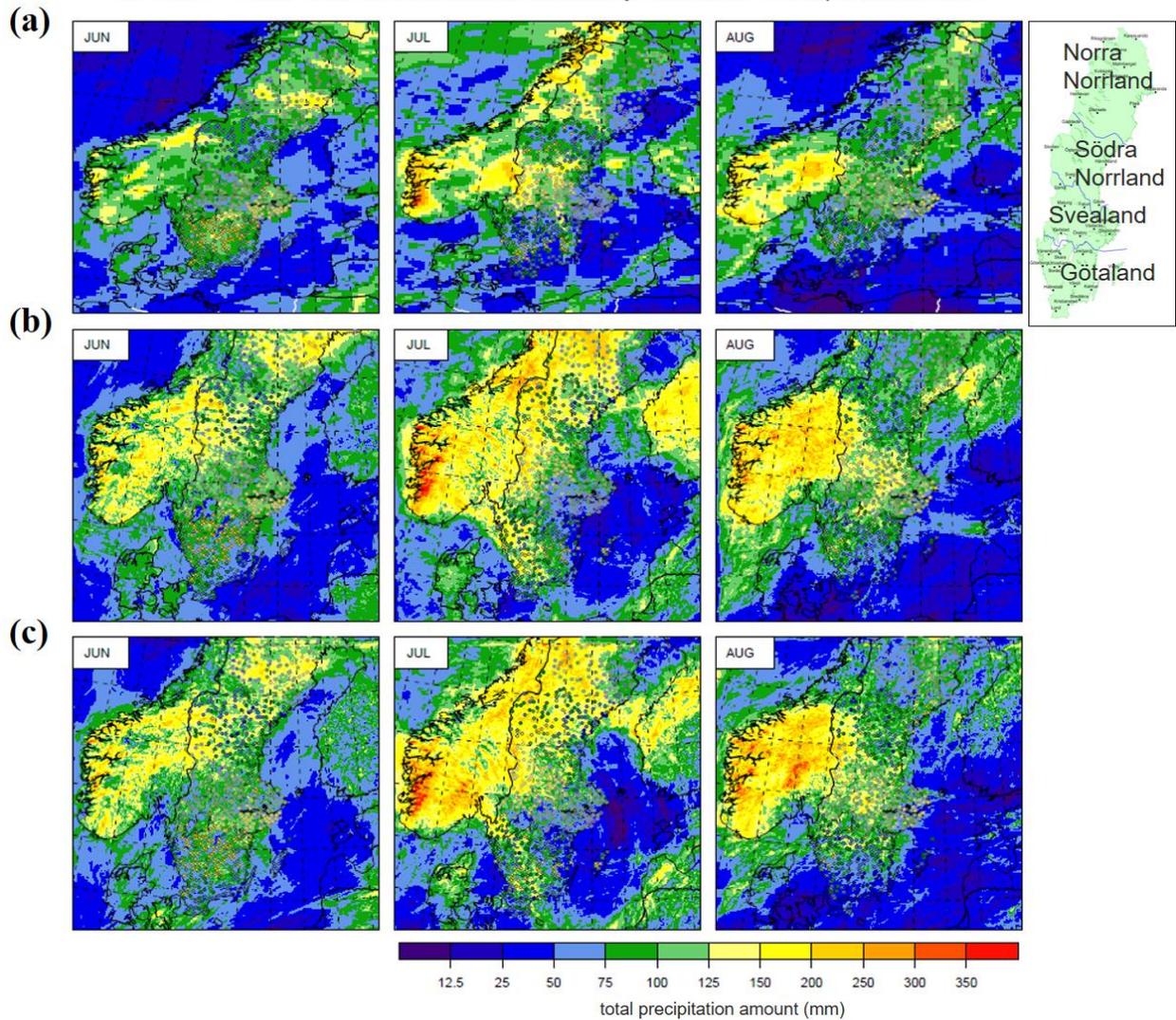


Figure S1. Monthly accumulated precipitation (mm) maps for summer months (JJA) in 2012: computed precipitation fields from different configurations of COSMO-CLM compared to observations at meteorological stations shown as circles filled with corresponding colour of observed precipitation: (a) COSMO-CLM on 0.11 degrees ("011"), (b) COSMO-CLM on 0.025 degrees with parameterised convection ("0025_Tiedtke") and (c) COSMO-CLM on 0.025 degrees with convection-permitting configuration ("0025_convper"), as used for the CMAQ high resolution domain. Observation data is based on precipitation measurements at 1804 stations of the Swedish meteorological network from SMHI. Inset in the top right corner shows the definition of the four regions of Sweden with blue border lines.

Probability distribution of differences in total precipitation SMHI station – CCLM 2012

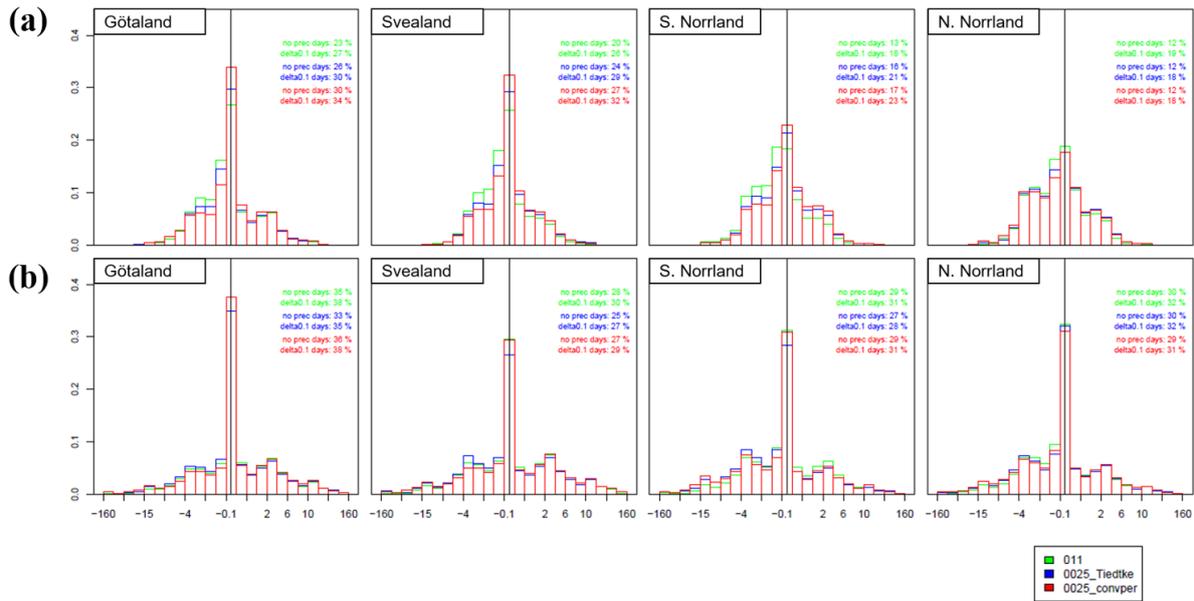


Figure S2. Probability distribution of the differences in daily precipitation sums (mm d^{-1}) between the SMHI station observations and COSMO-CLM with different configurations (“011”, “0025_Tiedtke”, and “0025_convper”) in the four regions of Sweden (from left to right: Götaland, Svealand, Södra Norrland, Norra Norrland): (a) for the winter months (JFD) of 2012 and (b) for the summer months (JJA) of 2012. The percentage fraction of days with zero difference between model and observation (“no prec.” days) and the percentage fraction of days with difference of $\pm 0.1 \text{ mm d}^{-1}$ (“delta0.1” days) is indicated in the plots for each model configuration.

Probability distribution of differences in total precipitation SMHI station – CCLM Götaland 2012

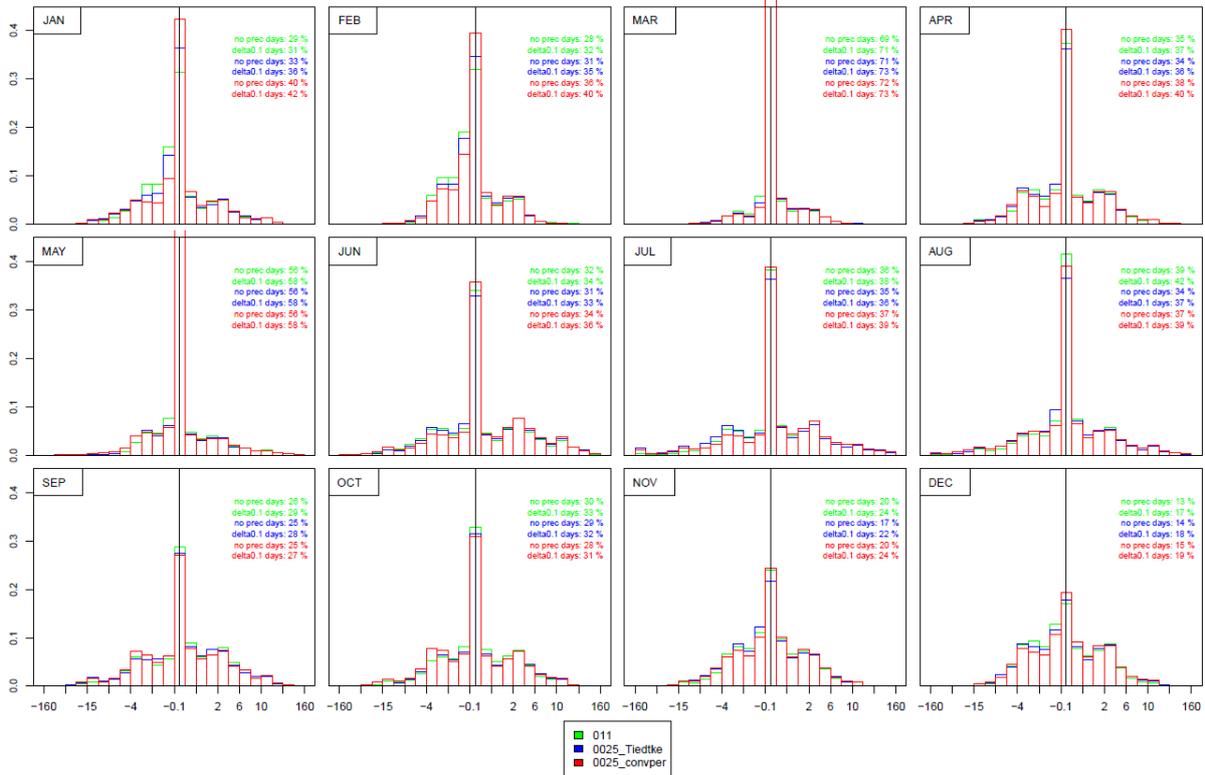
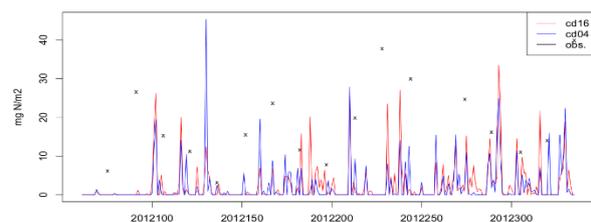
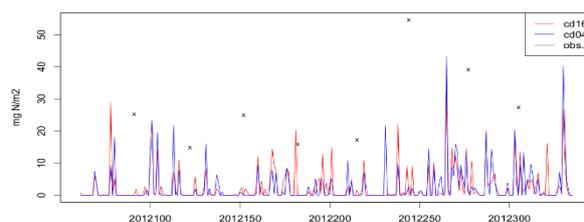


Figure S3. Probability distribution of the differences in daily precipitation sums (mm d^{-1}) between the SMHI station observations and COSMO-CLM with different configurations (“011”, “0025_Tiedtke”, and “0025_convper”) in Götaland for the months of 2012. The percentage fraction of days with zero difference between model and observation (“no prec. days”) and the percentage fraction of days with difference of $\pm 0.1 \text{ mm d}^{-1}$ (“delta0.1 days”) is indicated in the plots for each model configuration

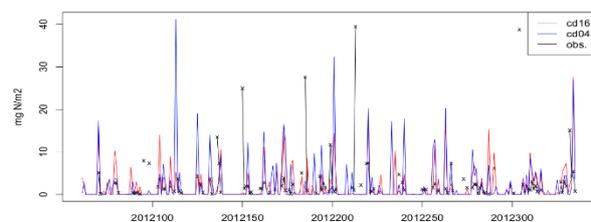
(a) Anholt, DK0008R



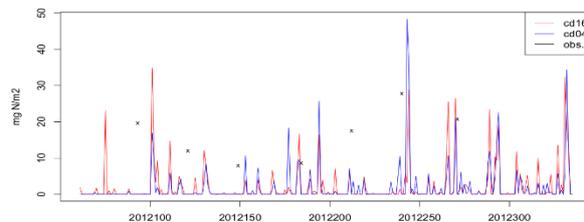
(b) Vavihill, SE0011R



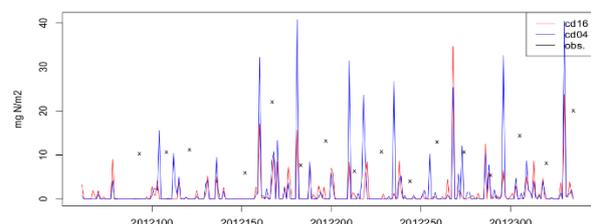
(c) Diabla Gora, PL0005R



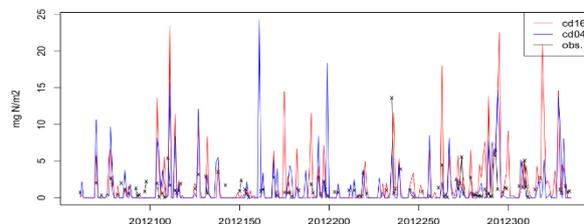
(d) Aspvreten, SE0012R



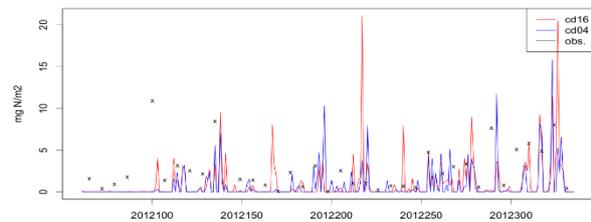
(e) Keldsnor, DK0005R



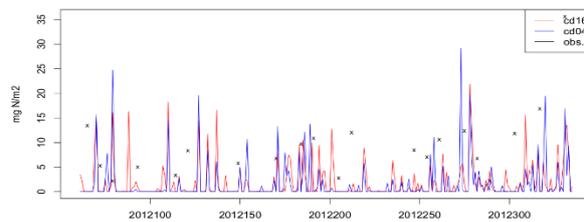
(f) Lahemaa, EE0009R



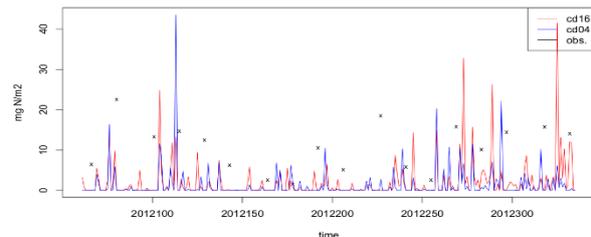
(g) Hailuoto II, FI0053R



(h) Rucava, LV0010R



(i) Vilsandi, EE0011R



(j) Sepstrup Sande, DK0022R

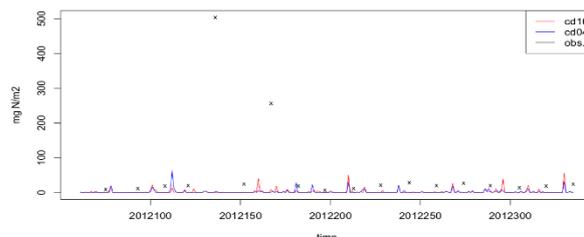
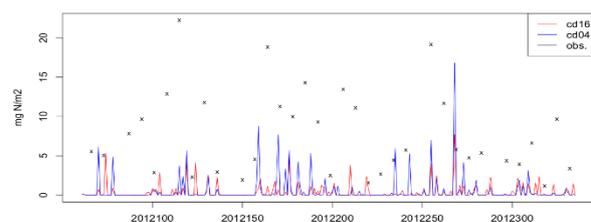
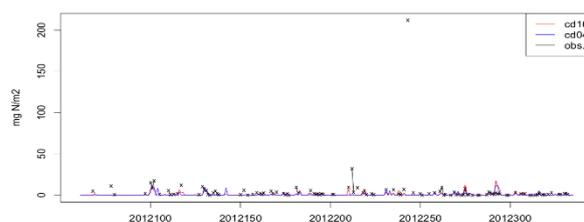


Figure S4. Comparison of modelled wet deposition of nitrate as daily sums ($\text{mg(N)} \text{m}^{-2} \text{d}^{-1}$) from the 16-km resolution grid (red) and 4km-resolution grid (blue) against observed daily sums of nitrate wet deposition (black crosses) at regional background stations around the Baltic Sea from the EMEP monitoring network: (a) Anholt, DK0008R, (b) Vavihill, SE0011R, (c) Diabla Gora, PL0005R, (d) Aspvreten, SE0012R, (e) Keldsnor, DK0005R, (f) Lahemaa, EE0009R, (g) Hailuoto II, FI0053R, (h) Rucava, LV0010R, (i) Vilsandi, EE0011R, and (j) Sepstrup Sande, DK0022R. Comparison time period: 1 March to 30 November 2012.

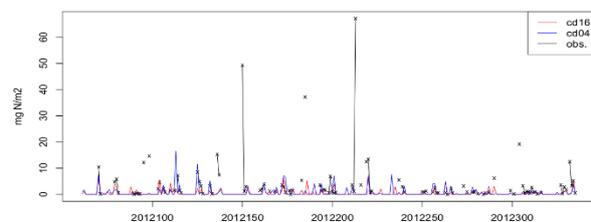
(a) Zingst, DE0009R



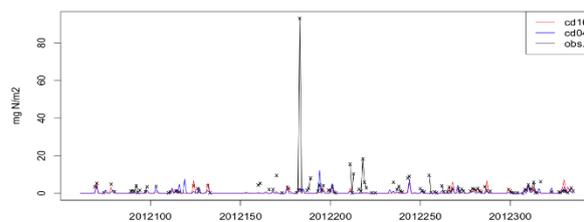
(b) Råö, SE0014R



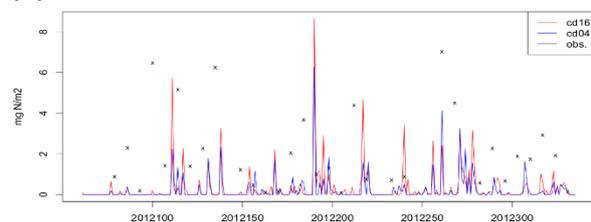
(c) Diabla Gora, PL0005R



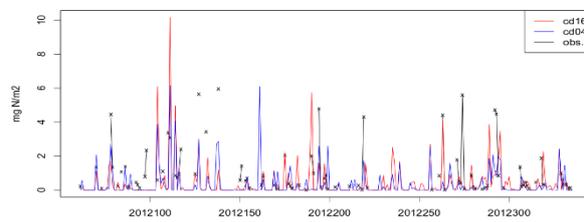
(d) Leba, PL0004R



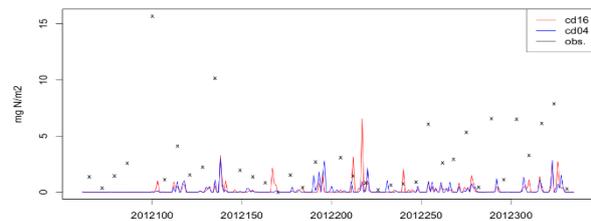
(e) Ahtari, FI0004R



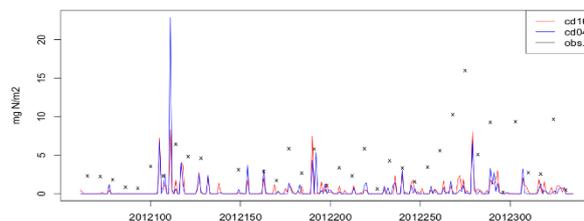
(f) Lahemaa, EE0009R



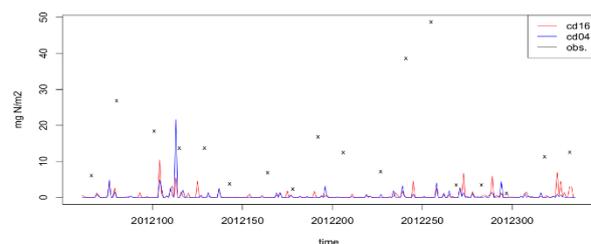
(g) Hailuoto II, FI0053R



(h) Virolahti II, FI0017R



(i) Vilsandi, EE0011R



(j) Preila, LT0015R

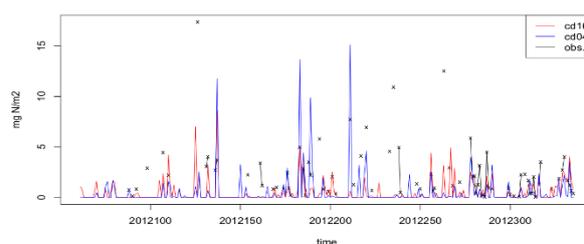


Figure S5. Comparison of modelled wet deposition of ammonium as daily sums ($\text{mg(N) m}^{-2} \text{d}^{-1}$) from the 16-km resolution grid (red) and 4km-resolution grid (blue) against observed daily sums of nitrate wet deposition (black crosses) at regional background stations around the Baltic Sea from the EMEP monitoring network: (a) Zingst, DE0009R, (b) Råö, SE0014R, (c) Diabla Gora, PL0005R, (d) Leba, PL0004R, (e) Ähtäri, FI0004R, (f) Lahemaa, EE0009R, (g) Hailuoto II, FI0053R, (h) Virolahti II, FI0017R, (i) Vilsandi, EE0011R, and (j) Preila, LT0015R. Stations correspond to those in Table 5. Comparison time period: 1 March to 30 November 2012.

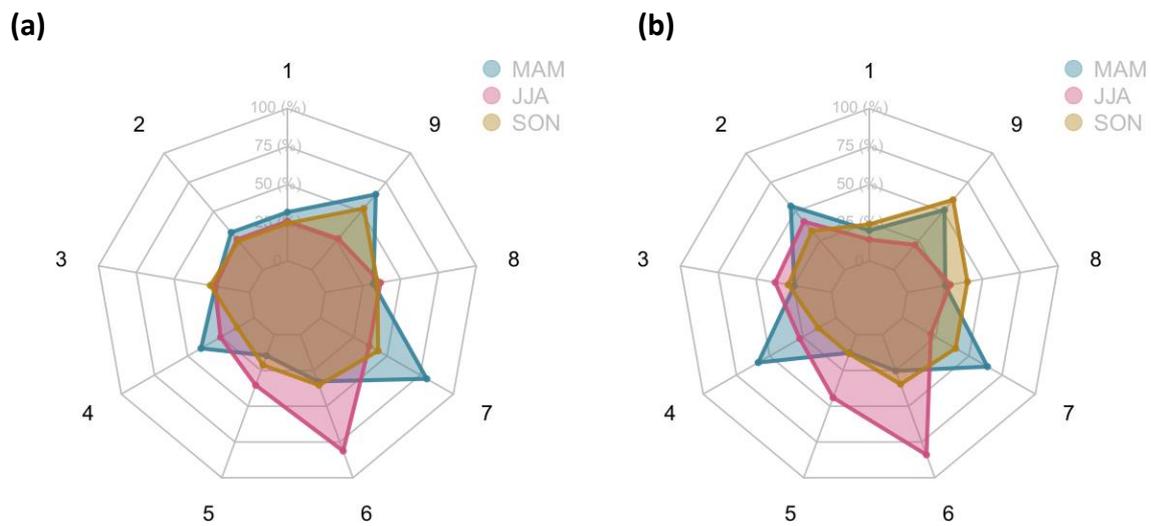


Figure S6. Spider charts of the ratio between modelled and observed seasonal averages: (a) wet deposition of nitrate and (b) wet deposition of ammonium, for spring (MAM), summer (JJA) and autumn (SON). Stations indicated by numbers 1: Zingst, DE0009R; 2: Lahemaa, EE0009R; 3: Ähtäri, FI0004R; 4: Virolahti II, FI0017R; 5: Hailuoto II, 6: Preila, LT0015R; 7: Leba, PL0004R; 8: Diabla Gora, PL0005R; 9: Råö, SE0014R. The maximum (100 %) corresponds to a ratio of 1.5.

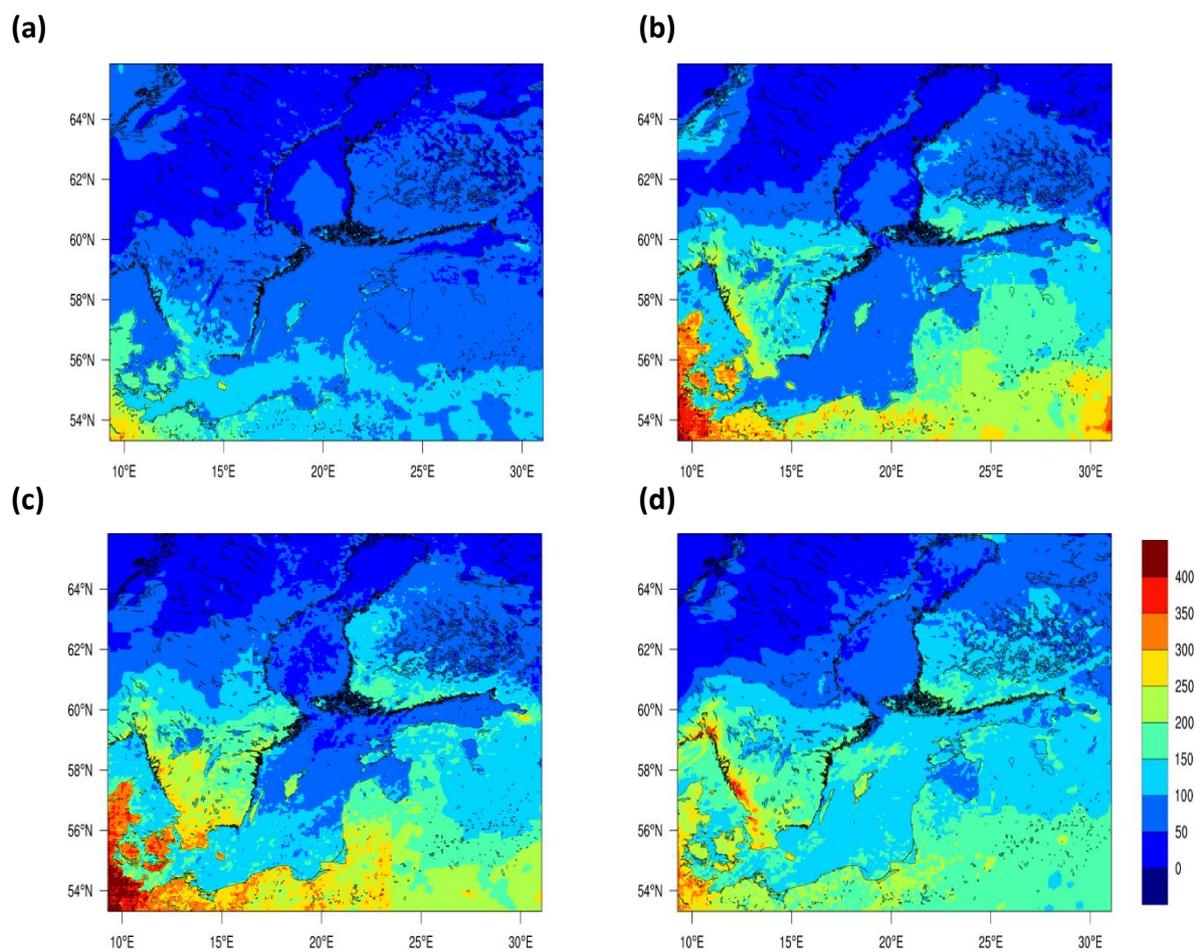


Figure S7. Present-day (2012) seasonal sums of total nitrogen deposition (in mg(N) m^{-2}) from the CMAQ run with all emissions: (a) in winter (JFD), (b) in spring (MAM), (c) in summer (JJA), and (d) in autumn (SON).

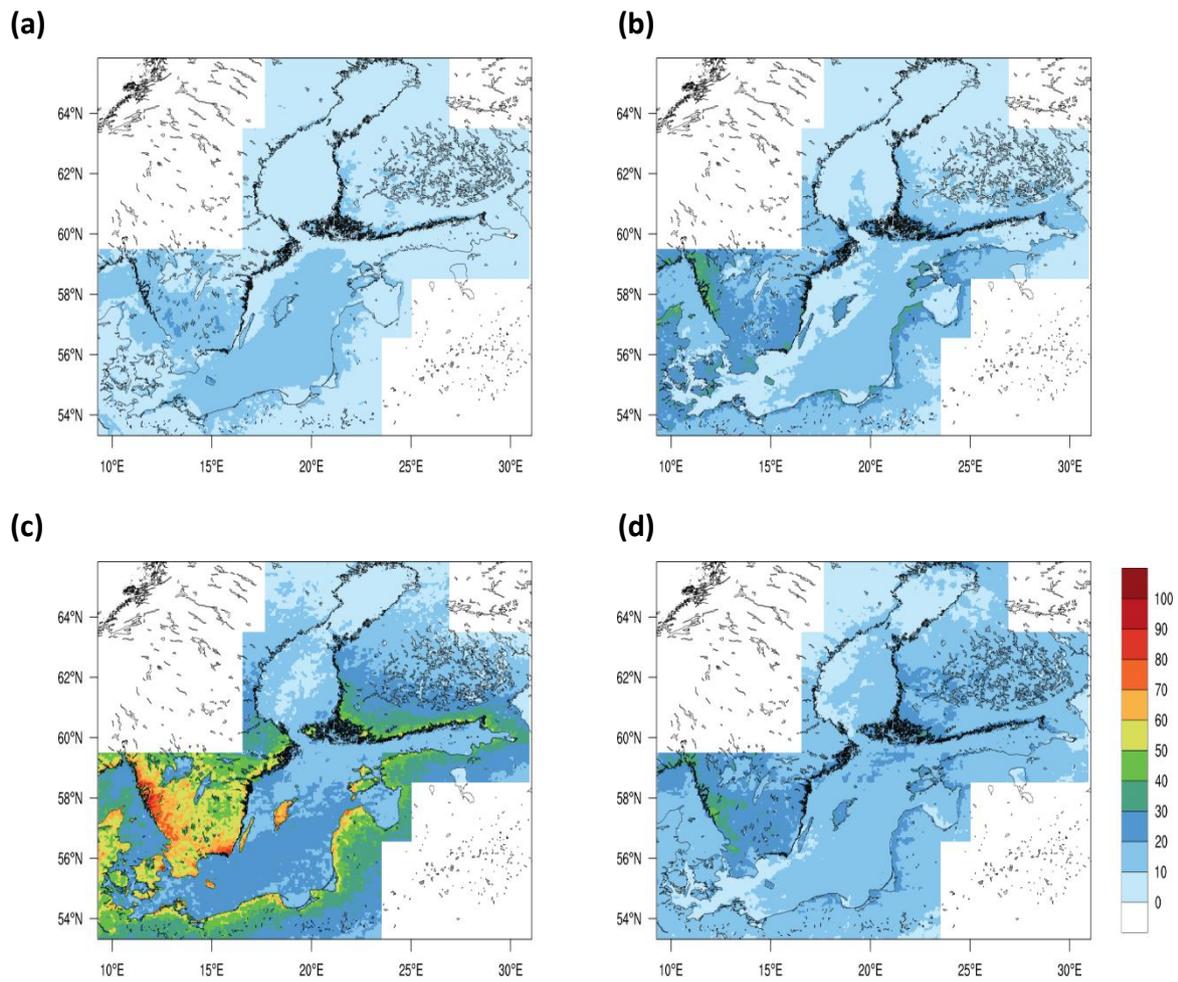


Figure S8. Present-day (2012) absolute ship contribution to the seasonal sums of nitrogen deposition (in mg(N) m^{-2}): (a) in winter (JFD), (b) in spring (MAM), (c) in summer (JJA), and (d) in autumn (SON). Maps only show results for the high-resolution area.

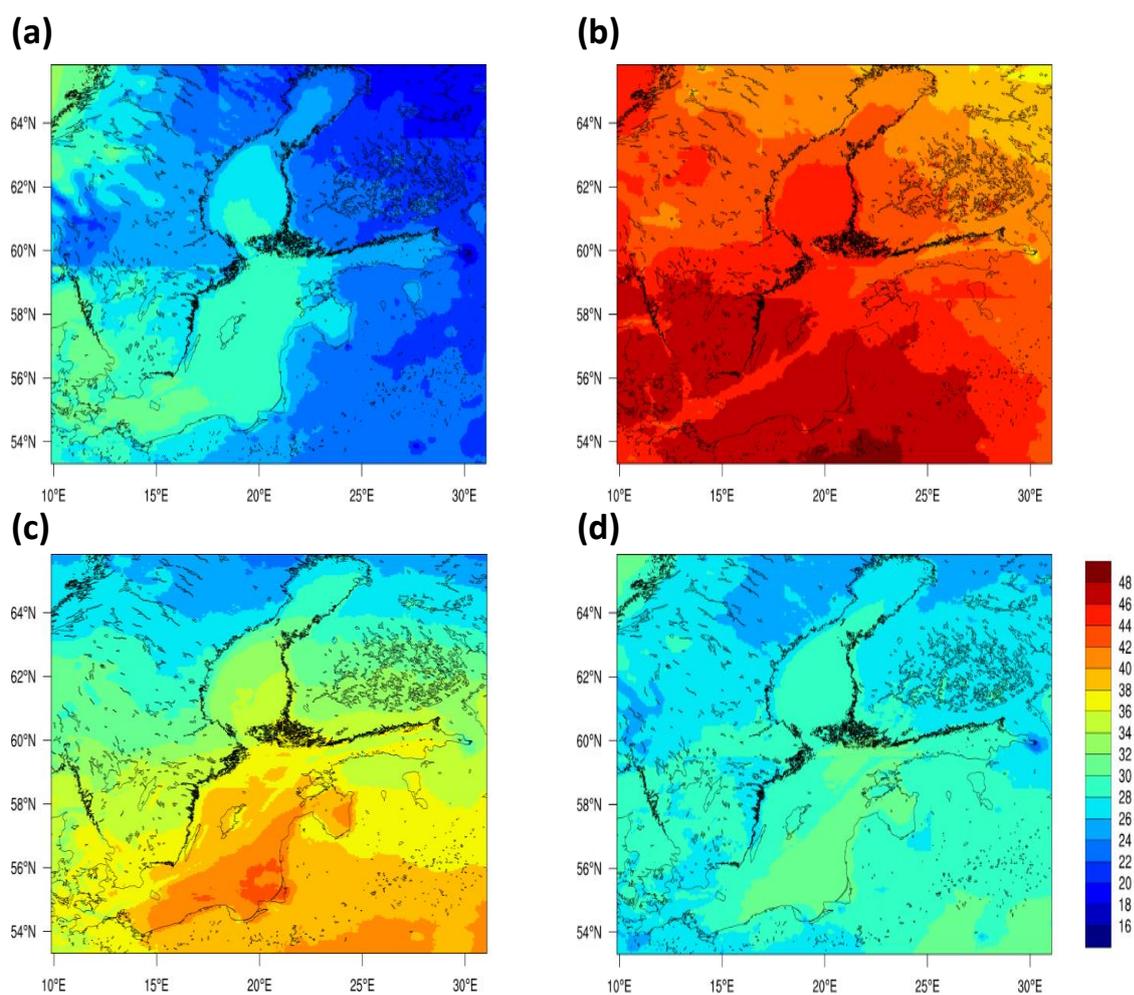


Figure S9. Present-day (2012) seasonal average of the daily maximum O₃ concentration (in ppbv) in the Baltic Sea region from the CMAQ run with all emissions: (a) mean of winter months (JFD), (b) mean of spring months (MAM), (c) mean of summer months (JJA), and (d) mean of autumn months (SON).

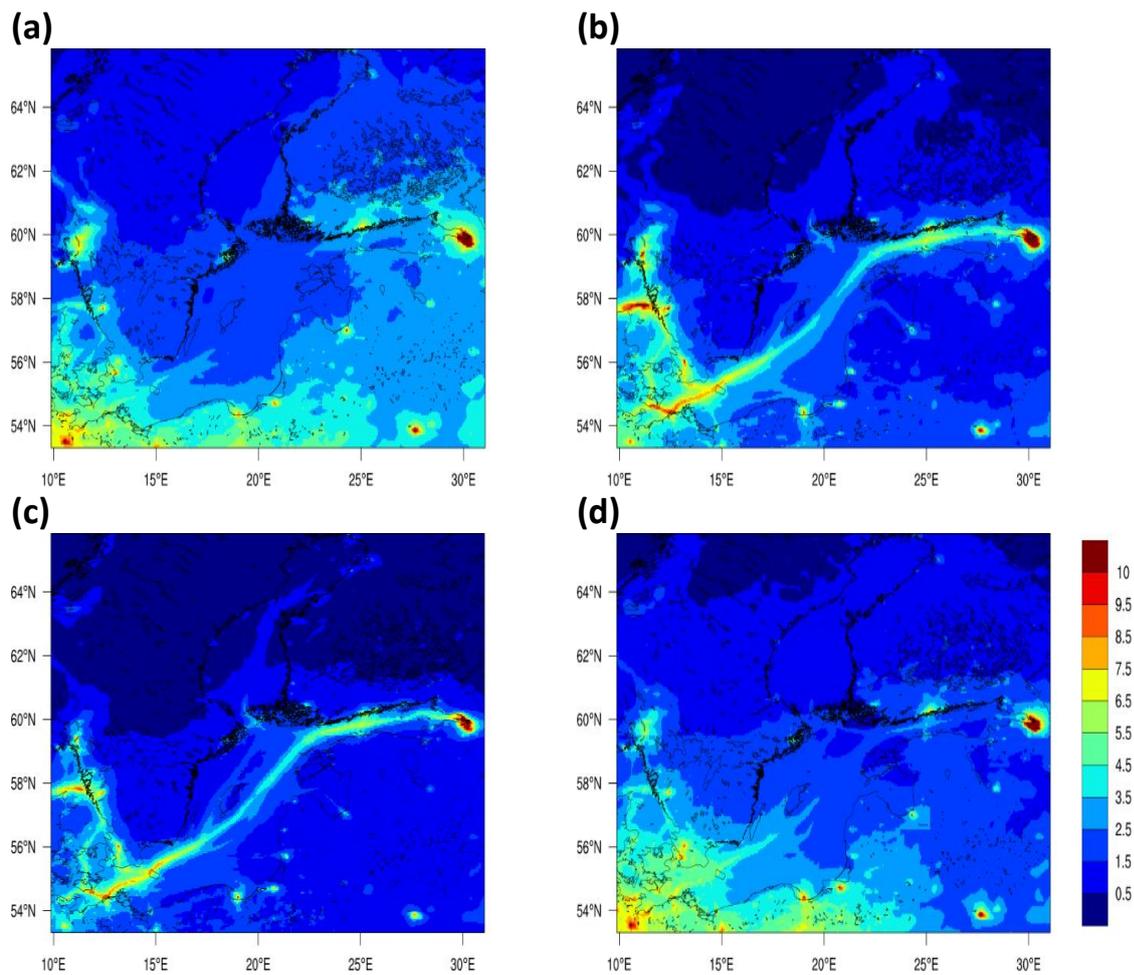


Figure S10. Present-day (2012) seasonal average concentration of NO₂ (in ppbv) in the Baltic Sea region from the CMAQ run with all emissions: (a) mean of winter months (JFD), (b) mean of spring months (MAM), (c) mean of summer months (JJA), and (d) mean of autumn months (SON).

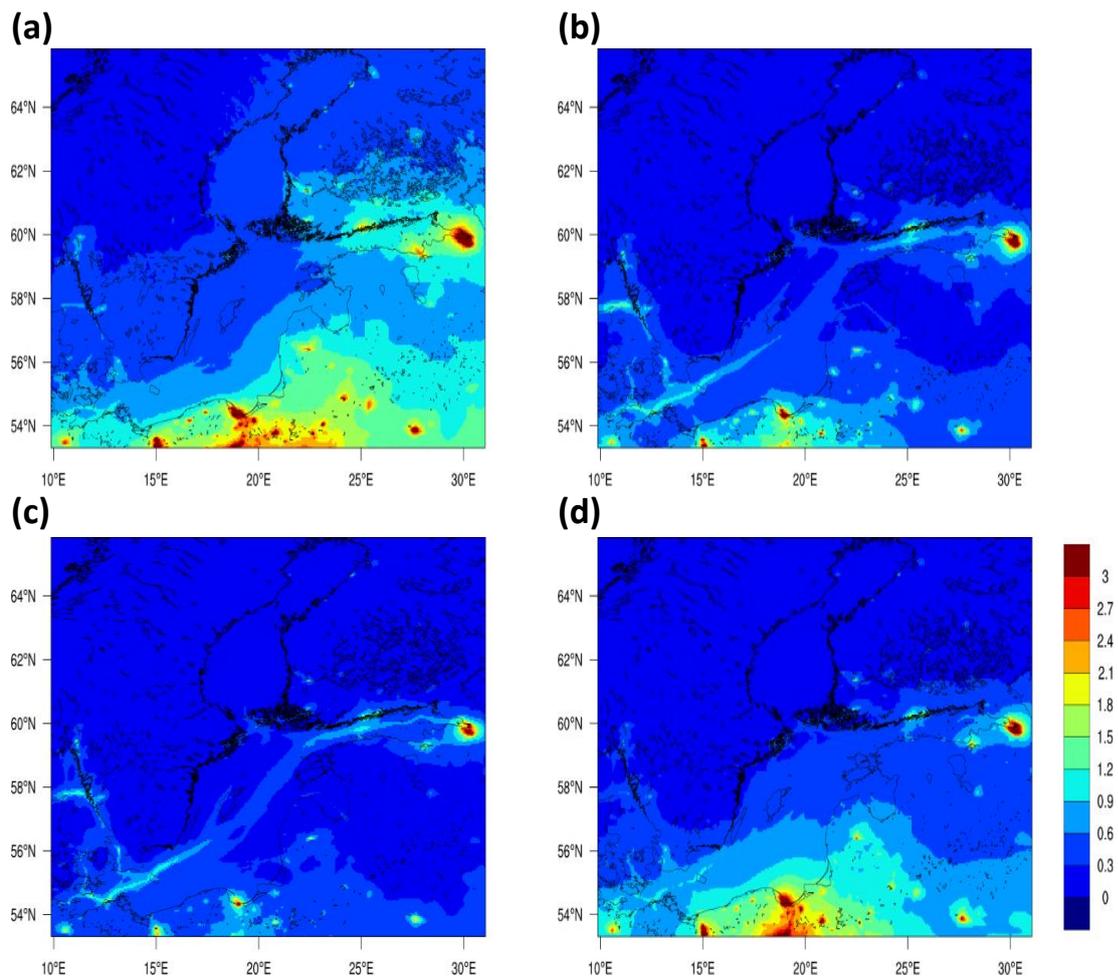


Figure S11. Present-day (2012) seasonal average concentration of SO₂ (in ppbv) in the Baltic Sea region from the CMAQ run with all emissions: (a) mean of winter months (JFD), (b) mean of spring months (MAM), (c) mean of summer months (JJA), and (d) mean of autumn months (SON).

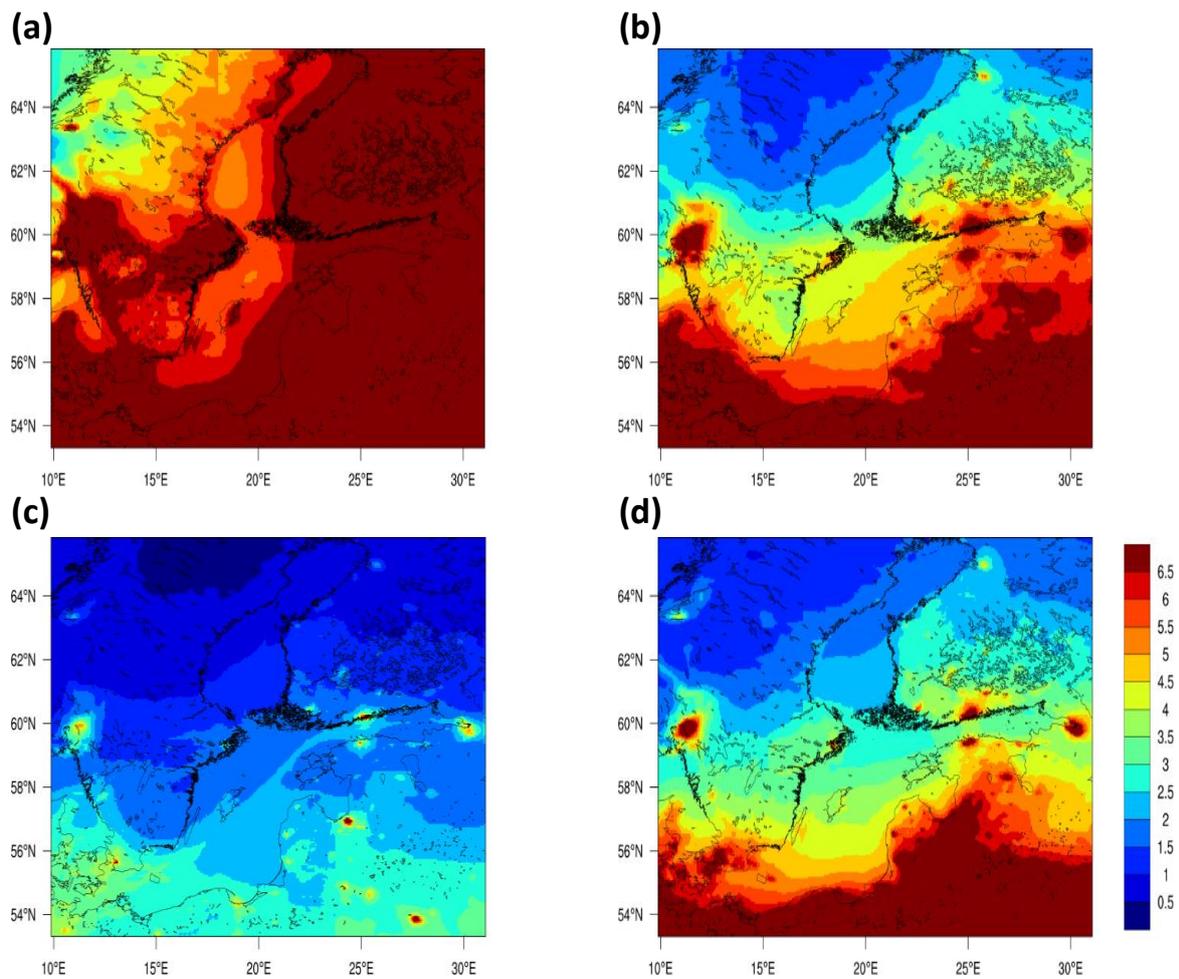


Figure S12. Present-day (2012) seasonal average concentration of PM_{2.5} (in $\mu\text{g m}^{-3}$) in the Baltic Sea region from the CMAQ run with all emissions: (a) mean of winter months (JFD), (b) mean of spring months (MAM), (c) mean of summer months (JJA), and (d) mean of autumn months (SON).

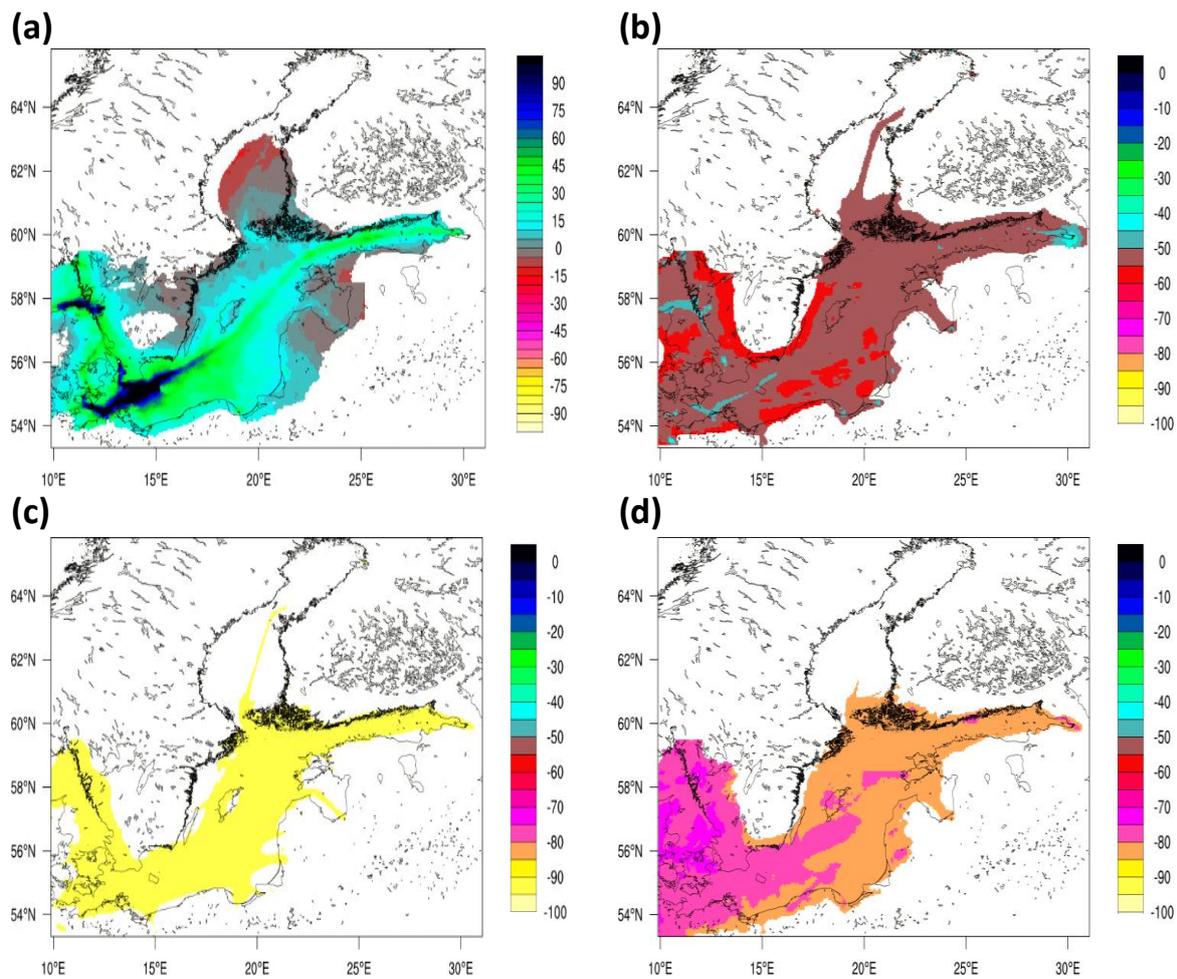


Figure S13. Future (2040) change of the ship-related contribution in summer (JJA) in percent compared to 2012, given as rel. difference between the ship contribution from the “NoNECA 2040” simulation and the ship contribution from the present-day simulation: (a) daily maximum O₃, (b) NO₂, (c) SO₂, (d) PM_{2.5}. Not coloured (empty) areas indicate grid cells with ship contribution in “BAU 2040” of less than 1.0 ppbv, 0.1 ppbv, 0.01 ppbv, 0.005 μg m⁻³, for daily max. O₃, NO₂, SO₂, PM_{2.5}, respectively. Ship-related contribution only shown for the high-resolution area. Note the different scale for daily max. O₃ (from -100 % to 100 %).

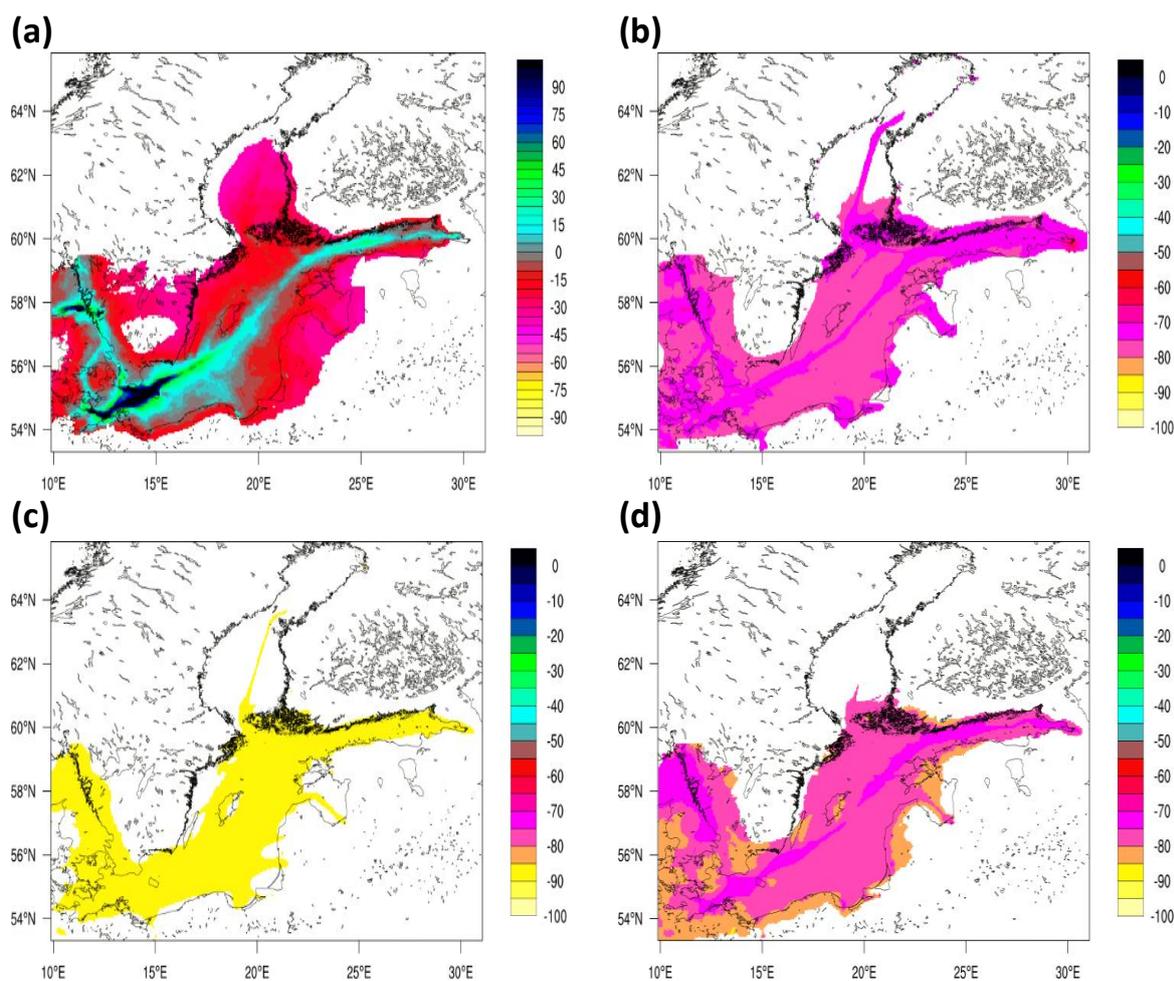


Figure S14. Future (2040) change of the ship-related contribution in summer (JJA) in percent compared to 2012, given as rel. difference between the ship contribution from the “EEDI 2040” simulation and the ship contribution from the present-day simulation: (a) daily maximum O₃, (b) NO₂, (c) SO₂, (d) PM_{2.5}. Not coloured (empty) areas indicate grid cells with ship contribution in “BAU 2040” of less than 1.0 ppbv, 0.1 ppbv, 0.01 ppbv, 0.005 $\mu\text{g m}^{-3}$, for daily max. O₃, NO₂, SO₂, PM_{2.5}, respectively. Ship-related contribution only shown for the high-resolution area. Note the different scale for daily max. O₃ (from -100 % to 100 %).

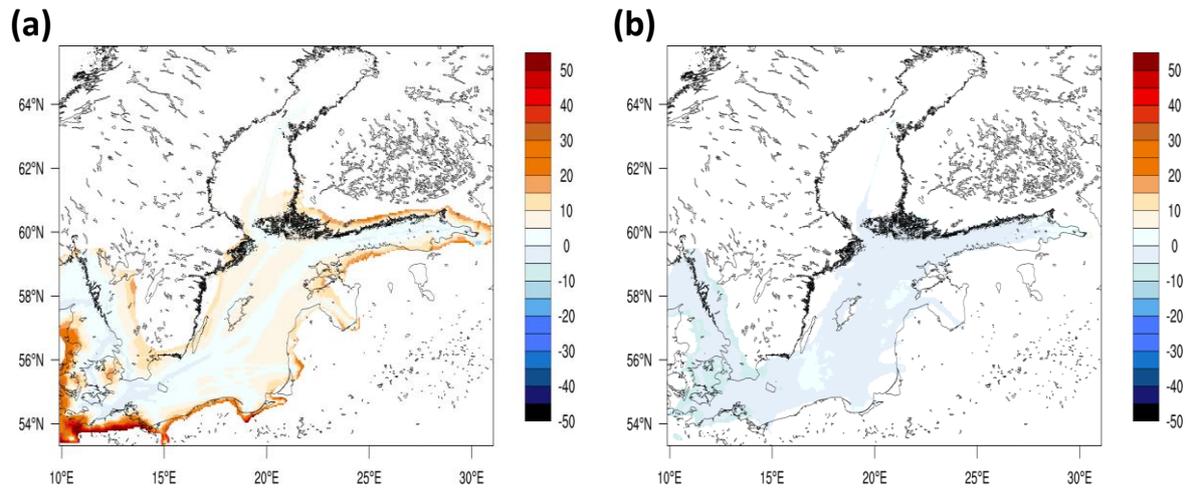


Figure S15. Effect of reduced land-based emissions (in percent) on the future ship contribution of (a) NO₂ and (b) SO₂ in summer (JJA) 2040 in the Baltic Sea region. Not coloured (white) areas indicate grid cells with ship contribution in the scenario “BAU 2040” of less than 0.1 ppbv for NO₂ and less than 0.01 ppbv for SO₂.

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