

First of all, we thank the reviewers for their thorough reviews and their comments that helped to improve the manuscript. We are grateful to reviewer 2 for reviewing both manuscripts at the same time. This gave us the opportunity to harmonize both parts and make better readable as two parts of a bigger project. We will now detail our answers to the reviewer's comments and list the changes we made.

Response to the comments of Reviewer 1

General comment

R: The paper is well written with a clear focus and relevance for the field of air quality and emission policy. The methods are straightforward and sound. The choice to look at relative changes in concentrations is appropriate, but at some locations the concentrations are very low, so I wonder whether the relative changes are meaningful there (e.g. impact of shipping on nitrate concentration in north eastern and south western part of North Sea, on ozone concentrations in English channel). A word on absolute concentrations would be helpful there.

A: We agree that at some locations, absolute concentration differences are very low and relative changes are not meaningful. Therefore, cutoff values were chosen below which the relative differences are not shown and the areas in the plots remain white. We added information about the cutoff value in the relevant figure captions. We don't show plots with the absolute differences for all substances because this would add too many plots to the paper that is already quite long. Absolute values are given for selected regions in Figs 6, 9, 12, A6, A7, A8.

R: Although I understand the choice of the authors to keep the anthropogenic emissions constant I would welcome an indication of the relative contribution to concentrations of shipping for 2030 in the discussion. In particular for ozone this would be relevant, as the chemical regime may change.

A: If European policies on the reduction of NO_x emission will be successful, we can expect lower NO_x emissions from land based sources in 2030 compared to today. This would mean that the relative contribution from shipping to NO_2 levels and to nitrate aerosol concentrations would be higher than shown here.

For ozone in summer, lower NO_x emissions from land would mean that even the area of the English Channel, which is currently VOC limited, could turn into a NO_x limited region. Additional NO_x emissions from ships would then enhance ozone where it is now reduced. This would be the case in the English Channel in the No ECA and the ECA SCR 21 scenarios. We discussed this already briefly in

the text at the end of the conclusions. We now added a small paragraph to this.

Detailed comment

R: P 11330 I 4-6: Grammatical construction of sentence is incorrect, which makes it difficult to read, please rephrase

A: Has been rephrased.

R: P 1135: Mention that the impact of shipping is determined by comparing a simulation including both shipping and other anthropogenic emissions to a simulation excluding the shipping emissions. Would you need to be concerned about nonlinearities by completely shutting down shipping emissions instead of looking at the impact of 10% reduction?

A: We added a paragraph on the method in the beginning of section 4.2. We tested possible nonlinearities in other applications by shutting down emissions from one sector by different percentages and found that the effects were very small. Nevertheless, nonlinearities might occur, because the entire chemical system is nonlinear. However, it is doubtful that the impact of shipping can better be quantified by upscaling the effects of a 10% emission change by a factor of 10.

R: Please describe what the white patches in the figures 2, 3, 5 and 7 mean, also how I should interpret the red patches close to Denmark and North of Ireland (Figs 2,3)

A: In the white areas in Figs 2 and 3 shipping emissions were too low (or even 0, e.g. over land) to calculate sensible quotients for the scenario maps. Red patches north of Ireland and close to the German coast are a result of low emissions in the reference case, too. We now also turned these regions into white areas, where no values are given. In Figs 5 and 7 white regions also denote areas with too low values to show sensible concentration ratios. We now give the limit values in the Figure caption.

R: P 1136 I 23: I would rather say 50 %, since it is nearly 100 % over large areas.

A: Has been changed, although large parts of those regions have low absolute concentrations.

Technical issues

R: Fig 6: name of scenarios not consistent with main text. Why 2a instead of 2? Better to refer to ECA specification.

A: Has been changed.

R: Fig 10: it would be helpful if SO₂ and SO₄ would be indicated next to the respective figure instead of only in the caption

A: Has been changed.

Response to the comments of Reviewer 2

R: a) Page 11326, L23-24: I would be careful with the transport fuel efficiency statement. True, ship on average, is the most efficient mode of transport, but very large differences exist between various ship types. Large intercontinental container ships or oil tankers are drastically more efficient than RoPax ferries.

A: We modified the sentence accordingly.

R: b) Page 11327, L5-6 and L8-9: Repetition of 0.1% S fuel requirement. I would also say that the EU sulphur directive is relevant in this context.

A: We omitted the sentence in Line 8/9 and added references to the EU sulphur directive.

R: c) Page 11327, L15-24. Here the authors shed some light on the NO_x emission factors used in part 1, but it is not reported in part 1 at all. Also, IMO Marpol Annex VI Reg 13 Paragraph 7 sets the NO_x emission requirements for ships in ECAs and extends the Tier 1 limits to vessels built in 1990's. Was this considered in the current work?

A: The way the emission factors were derived from the GL test bed data and the emission functions themselves will be described in more detail in part 1 of these articles. It is not considered that some ships that were built in the 1990s already comply to Tier 1 limits. This is because this rule applies only to ships with bigger engines (> 5000 kW) and only if the technical specifications allow for such a retrofit. The latter information was not available to us. In addition, the largest part of the bigger ships that sail the North Sea were built after 2000. In a sensitivity run of the emission model we estimated the activities of ships with engines > 5000 kW that were built in 1990s (in number of ship movements) to be 1% of all movements. We therefore think, that the effect of Marpol Annex VI Reg 13 Paragraph 7 on the NO_x emissions in the North Sea is small.

R: d) Page 11328, L22-23: Here a reference is made to part 1 of the combined manuscript, but the necessary details are not there unless the authors significantly improve the description of the emissions part of the manuscript.

A: The description of the emission factors has been significantly extended in Part 1.

R: e) Page 11329, L17: Fleet renewal rate. How is the 2.5% replacement of the fleet done? Do the oldest vessels go first or is this done with random sampling of ships regardless of vessel age?

A: The oldest ships go first. This seems to be more reasonable than a random sampling, although it might be a bit too optimistic in terms of emissions reductions. Certainly, also newer ships will be put out of service and some older will still remain in service. However, we found it hard to put numbers to the fraction of old ships that might remain in service. We added a sentence that briefly explains this.

R: f) Page 11331: Scenarios for ECA LNG16 and ECA LNG21. I do not see emission factors for engines using LNG fuel (or specific consumption) reported anywhere in part1 or part2. These should be added, as well the discussion regarding methane slip from marine engines.

A: We added a paragraph on the construction of the LNG scenarios.

R: g) I agree with the statement made by Referee1 that land based emissions change, too. EMEP has made projections for 2030, which could be used for this purpose. The authors are actually quite optimistic with the reduction of land based emissions if they expect no growth between present date and 2030.

A: We added a brief discussion about the effects of changing land based emissions. See our answer to the comments of Referee 1. However, we disagree with the reviewer that keeping the land based emissions constant would be an optimistic scenario. In the RCP scenarios of the IPCC, OECD states expect a decrease in e.g. NOx emissions from 2010 until 2030 of 30% or more, the same holds for Sulphur emissions (see <http://tntcat.iiasa.ac.at/RcpDb/dsd?Action=htmlpage&page=compare>, last accessed 18 August 2015). EU expects a decrease in SO2 and NOx even under current legislation, by more than 50% between 2005 and 2050 (see Amann (Ed), The final policy scenarios of the EU clean air policy package, TSAP report 11, February 2014 http://www.iiasa.ac.at/web/home/research/researchPrograms/MitigationofAirPollutionandGreenhousegases/TSAP_11-finalv1-1a.pdf, last accessed 18 August 2015)

R: h) Page 11335, L17-19: Conversion rates are not the only factors increasing the concentrations, because also ship activity is higher during summer than in winter.

A: We now explain this in more detail.

R: i) Page 11337, L18-21: This "All ships in Tier3 by 2030" is not a very realistic scenario. I understand it sets the game with Maximum Feasible Reduction and the reductions available through drastic measures, but I do not believe for a second that this would happen in reality.

A: We agree, but as you pointed out, this scenario sets the scene for what could be possible and what would be reality in e.g. 2050 when most ships would be replaced by new ones (and assuming that the total transported volume will not drastically increase further).

R: j) Page 11341, Chapter 4.2.5 "PM2.5": The discussion of PM2.5 feels weak because it is not reported in part1 or part2 how the emission factors for PM are impacted by fuel sulphur content.

A: The emission factors for PM are now reported and discussed in Part 1 of the two papers. We describe the changes applied in the scenarios in section 2.3.

R: k) Page 11354 and Page 11357, Figures 6 and 9: These figures are an excellent idea how to describe the impact of scenarios.

A: We agree.

List of changes:

line 26-29

EU has implemented these rules in their directives 1999/32/EC (European Union, 1999) and 2012/33/EU (European Union, 2012) for the North and the Baltic Seas which are Sulphur Emission Control Areas (SECAs). This means that the fuel burned in these areas must not contain more than 0.1% sulphur (S) (until 31 Dec 2014 1.0% S).

was added

line 85

Older ships are replaced first.

was added

line 97-98

Tier III NO_x regulations apply only to new ships. Therefore, it needs some time until a considerable number of ships in the fleet will have reduced NO_x emissions.

was added

line 144-151 The LNG scenarios differ from the SCR scenarios in the following way: SO₂ emissions are zero while they are about 0.38 g/kWh for the SCR case. This a reduction by a factor of 10 compared to the base case and corresponds to a sulphur content of 0.1% S, an average fuel consumption of 200 g/kWh and a ratio of 95% of the total sulphur emitted as SO₂. Also PM emissions are set to zero in the LNG case. For SCR use, sulphate emissions are again reduced by a factor of 10 according to the sulphur reduction from 1.0% S to 0.1% S in the fuel. Other PM emissions are kept constant. NO_x emissions were the same for both, LNG and SCR scenarios. They differ in the implementation date of Tier III rules, only.

was added

line 224-226

Partly, they can be ascribed to seasonal differences in the emissions with higher shipping emissions in summer. Most of the differences in the concentrations are caused by atmospheric chemistry.

was added

line 235-239

To derive the contribution of ships to the selected pollutant concentrations two model runs, one including and one excluding shipping emissions, were performed. The difference is regarded as the contribution of ships to the individual pollutant. For the scenarios the difference between two model runs with different shipping emissions is regarded as the change in the contribution of ships in the respective scenario.

was added

line 429-432

Ozone might increase in regions where it was found to decrease in the scenarios No ECA and ECA SCR 16, namely in the English Channel. When this region, where ozone formation is currently VOC limited, turns into a NO_x limited region, NO_x emissions from shipping will enhance ozone concentrations in the entire study area.

was added

line 469-474

References to the EU sulphur directive were added

line 492-493

The reference was updated

Fig.2

No values are shown in grid boxes where the NO_x emissions from ships were below 0.5 t/a per grid cell of $24 \times 24 \text{ km}^2$.

was added

Fig.3

No values are shown in grid boxes where the SO_2 emissions from ships were below 0.5 t/a per grid cell of $24 \times 24 \text{ km}^2$.

was added

Fig.5

No values are shown in grid boxes where the contribution from shipping to the NO_2 concentrations in either the reference or the scenario case was below 0.05 $\mu\text{g}/\text{m}^3$.

was added

Fig.7

No values are shown in grid boxes where the contribution from shipping to the NO_3 concentrations in either the reference or the scenario case was below 0.005 $\mu\text{g}/\text{m}^3$.

was added

Fig.10

Labels for SO_2 and SO_4 were added.

Fig. 6, 9, 12, A6, A7, A8

Labels were changed according to the scenario names into No ECA, ECA SCR 16, ECA SCR21, ECA opt.

The impact of shipping emissions on air pollution in the Greater North Sea region. Part II: Scenarios for 2030

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Abstract. Scenarios for future shipping emissions in the North Sea have been developed in the framework of the Clean North Sea Shipping project. The effects of changing NO_x and SO₂ emissions were investigated with the chemistry transport model CMAQ for the year 2030 in the North Sea area. It has been found that, compared to today, the contribution of shipping to the NO₂ and

O₃ concentrations will increase due to the expected enhanced traffic by more than 20% and 5%, respectively, by 2030 if no regulation for further emission reductions will be implemented in the North Sea area. PM_{2.5} will decrease slightly because the sulphur contents in ship fuels will be reduced as international regulations foresee. The effects differ largely between regions, seasons and date of the implementation of stricter regulations for NO_x emissions from new built ships.

10 1 Introduction

Shipping is an important contributor to air pollution in coastal areas. More than 90% of the global trade is done with ships. The total global transport work by ships (in tonne miles) has been tripled since the mid 1980s (Smith et al., 2014), corresponding to an average grow rate of 4% p.a., and the forecasts for the future are in the same order of magnitude (Smith et al., 2014). The North Sea is one of the areas with the highest ship densities in the world. Europe's three biggest harbours in Rotterdam, Hamburg and Antwerp are located in the North Sea region. At any time about 3000 ships are sailing in the North Sea (Aulinger et al., 2015). This steady increase in number and size of ships leads to an increasing contribution of ships to air pollution in North Sea coastal areas (Matthias et al., 2010; Hammingh et al., 2012; Jalkanen et al., 2012; Aulinger et al., 2015). Compared to other

20 modes of transport like trucks or trains, big container ships or tankers are very efficient in terms of fuel use per ton mile. However, NO_x , SO_2 and PM emissions are comparably high because of less strict regulations for the emissions of these pollutants from ships. This problem has already been recognized years ago leading to stricter regulations in some areas, the so called Emission Control Areas (ECAs). These regulations are the results of an agreement within the International Maritime
25 Organization (IMO) and they are laid down in MARPOL Annex VI (International Maritime Organisation, 2008). EU has implemented these rules in their directives 1999/32/EC (European Union, 1999) and 2012/33/EU (European Union, 2012) for the North and the Baltic Seas which are Sulphur Emission Control Areas (SECAs). This means that the fuel burned in these areas must not contain more than 0.1% sulphur (S) (until 31 Dec 2014 1.0% S). If fuels with higher sulphur content are
30 used, the exhaust gas has to be cleaned until it doesn't contain more sulphur than exhaust gas from a low-sulphur fuel. As a consequence, ships use low sulphur fuels or installed scrubbers on board that clean the exhaust gas from sulphur and other contaminants. Outside SECAs, the allowed sulphur content in ship fuels is currently at 3.5%, but it will be reduced to 0.5% in 2020 or 2025 at the latest. The exact date will be decided in 2018 when the availability of ship fuel containing less than 0.5%
35 S will be reviewed.

Nitrogen oxide emissions from ships are also regulated in MARPOL Annex VI. Since the year 2000 the NO_x limits for ships built after 1 January 2000 are 17 g/kWh NO_x (Tier I regulation). In 2010 the second step (Tier II) of the NO_x regulations came into force with an emission factor of 14.4 g/kWh NO_x for new ships. The third step (Tier III), when NO_x limits will be further reduced to 3.5
40 g/kWh NO_x , is planned for 2016 for the ECAs around North America. However, this will only be applied to new ships sailing in designated Emission Control Areas while Tier I and Tier II represent global limits. The North Sea and the Baltic Sea are in discussion to become such an ECA for NO_x . However, Tier III rules will only be valid for ships built after the designation date (International Maritime Organisation, Marine Environment Protection Committee, 2014). Until now, it is unclear
45 when this date will be.

In the European project Clean North Sea Shipping (CNSS) different technologies capable of reducing air emissions from ships in the North Sea were investigated. Among them are scrubbers that reduce sulphur emissions, catalysts that reduce NO_x emissions and the use of alternative fuels like liquefied natural gas (LNG). In order to estimate the effect of these technologies and of legislation
50 on NO_x and SO_2 emissions from ships, emission scenarios were developed for the year 2030. These scenarios consider the same development of the world fleet and different developments in legislation and the use of alternative fuels. The basis is a detailed emission inventory for the year 2011 which is built upon AIS (Automatic Identification System) ship positions and a detailed ship characteristics data base (Aulinger et al., 2015). The scenarios are implemented as modified emission inventories
55 for the year 2030. The inventories serve as input for the chemistry transport model CMAQ that is setup for the North Sea region. CMAQ calculates transport and transformation of the emitted pollu-

tants and finally yields concentration maps that illustrate the impact of shipping emissions on the air quality in the North Sea region.

2 Ship emission inventories

60 2.1 Reference emissions

The basis for the ship fleet and the ship movements on the North Sea is a data set with AIS positions of ships for the entire year 2011 combined with a ship characteristics data base that includes all ships given in the AIS data set. The data is used to calculate the energy demand of individual ships depending on the installed engine and their actual velocity. From this, fuel use as well as
65 NO_x, SO₂, CO₂, CO, Hydrocarbon (HC), and Particulate Matter (PM) emissions are calculated with load-dependent emission factors for the different species. For the first time, load dependent emission factors resulting from test bed measurements of about 250 different ship engines were used to calculate a ship emission inventory. For the details, the reader is directed to the accompanying paper by Aulinger et al. (2015).

70 2.2 Scenario description

The purpose of scenarios is to describe plausible and possible future developments. Scenarios are often used to describe the boundaries of possible future situations, e.g. a worst case and a best case. In our study we decided to create scenarios that describe the future development of policy and technology regarding exhaust gas emissions from ships in the North Sea area. We adopted the methodology
75 described in (Eyring et al., 2005) for our scenarios and distinguish between traffic demand and future technological and legislative developments. However, because we focus on the implementation of a NO_x emission control area in the North Sea, we take only one scenario for the fleet development into account as a basis as it is described in publications from IMO (Buhaug et al., 2009; Smith et al., 2014) and Det Norske Veritas (Det Norske Veritas, 2012). Taking multiple possible developments
80 of the world trade into account would add too much complexity to the scenarios.

In brief, our fleet development scenario assumes an increase in the number of bigger ships while the number of smaller ships decreases in the North Sea area. This leads to an increase in ship number by 1.0% p.a. and an increase in transported cargo of 2.5% p.a. Additional to this increase in ship number it is assumed that per year 2.5% of all ships are replaced by new ones, no matter of what size
85 they are. **Older ships are replaced first**. The main techniques under investigation are Liquefied Natural Gas (LNG) as an alternative fuel for shipping and end-of-the-pipe technologies like scrubbers and Selective Catalytic Reduction (SCR) to reduce sulphur dioxide and nitrogen oxide emissions. The main drivers for changes in the use of ship fuels and in the amount of emissions to air are on the one hand regulations, and here mainly what is written in MARPOL Annex VI (International
90 Maritime Organisation, 2008), and on the other hand the price of different fuels. Therefore, the main

scenarios include strict and less strict legislations as one axis and the price of LNG compared to Marine Gas Oil (MGO) or Heavy Fuel Oil (HFO) as the second axis. Some regulations in MARPOL Annex VI (those related to NO_x emissions) are only valid for newly built ships after a certain date, depending on the region where the ECA is located. The earliest date when Tier III NO_x regulations
95 will come into force is 1 January 2016. For the North Sea and the Baltic Sea, it is likely that Tier III NO_x regulations will be implemented significantly later than 2016.

Tier III NO_x regulations apply only to new ships. Therefore, it needs some time until a considerable number of ships in the fleet will have reduced NO_x emissions. Those regulations related to the sulphur content in ship fuels apply to all ships, and should have immediate effects on the total emissions
100 of sulphur oxides. To particularly take into account the long term effects of new ships following Tier III regulations, the year 2030 is used in the scenarios as target year. The development of the world fleet until 2030 compared to the reference year 2011 is considered.

These drivers are combined into six scenarios that can be arranged in a coordinate system with legislation on the x-axis and LNG price on the y-axis (see Fig. 1). Different implementation dates for
105 NO_x Tier III rules (2016 and 2021) are chosen.

The stories behind these scenarios can be described as follows:

Scenario No ECA

The global economy suffers from low GDP growth rates and in order to avoid additional costs for the shipping industry some regulations will not be implemented (global SO_x limit, NO_x limits in
110 ECAs). This can be considered as the worst case scenario, however 0.1% S in fuels in ECAs will still be implemented.

Scenarios ECA SCR 16 and ECA SCR 21

All regulations currently given in MARPOL Annex VI will be in force. The global sulphur limit of 0.5% S in fuel will be in force by 2020, in ECAs a sulphur limit of 0.1% S will be implemented
115 since 2015. A NO_x emission control area will be implemented in the North and Baltic Seas. Two different years, 2016 and 2021, are considered as implementation dates. LNG is expensive and the LNG infrastructure is not built up to provide LNG to many ships. Therefore ship owners will prefer low sulphur fuels and catalysts (SCR) or exhaust gas recirculation systems (EGR) to comply with the rules. Some will use scrubbers only, if they do not have to follow the Tier III regulations (older
120 ships).

Scenario ECA LNG 16 and ECA LNG 21

The legislation is the same as in scenarios ECA SCR 16 and ECA SCR 21 but LNG will be the cheaper solution to comply with the rules. In 2030, about 6000 ships in the North Sea will run on LNG. Ships that sail more than 50% of the time in the North and Baltic Seas will preferably use
125 LNG. Some newer ships will also be retrofitted with LNG engines.

Scenario ECA opt

This is built on scenario ECA SCR but it assumes that the strict rules for NO_x emissions for new-buildings will also apply to older ships in 2030. They will then be retrofitted with exhaust gas cleaning systems in order to follow these rules. This is regarded as the best case scenario and illustrates the reduction potential. However, exhaust gas cleaning systems increase the fuel consumption, so the fuel use in this scenario will be higher than in the previous scenarios in which only parts of the ships are equipped with exhaust gas cleaning.

2.3 Future shipping emissions

The emission inventories that were constructed as input for the CMAQ model were developed from the ship emission inventory for 2011 which is based on AIS data and ship characteristics data. First, the fleet development was applied. Then, the new emissions were calculated by using modified emission factors for the specific emissions of the ships. All emission factors are given in g/kWh for the different substances under investigation, they have been reduced depending on the scenario and taking into account the age of the different ships in the scenarios ECA SCR and ECA LNG. In particular, in all scenarios it has been taken into account that a fraction of the older ships, that do not have to follow any of the Tier rules for NO_x emissions, are taken out of service until 2030 and will be replaced by ships following Tier II. In areas where no AIS data was available (e.g. west of France), shipping emissions as given in the EMEP inventory were used.

The LNG scenarios differ from the SCR scenarios in the following way: SO_2 emissions are zero while they are about 0.38 g/kWh for the SCR case. This a reduction by a factor of 10 compared to the base case and corresponds to a sulphur content of 0.1% S, an average fuel consumption of 200 g/kWh and a ratio of 95% of the total sulphur emitted as SO_2 . Also PM emissions are set to zero in the LNG case. For SCR use, sulphate emissions are again reduced by a factor of 10 according to the sulphur reduction from 1.0% S to 0.1% S in the fuel. Other PM emissions are kept constant. NO_x emissions were the same for both, LNG and SCR scenarios. They differ in the implementation date of Tier III rules, only.

The change of the annual average emissions of NO_x and SO_x in comparison to the reference emissions of 2011 are given for the scenarios in Fig. 2 and Fig. 3, respectively. Regional differences in the emission changes are a result of ship types and ship sizes which undergo different temporal developments in fleet renewal. This depends on the actual age of the ships given in the ship characteristics data base.

3 Chemistry transport modeling

3.1 CMAQ

The CMAQ model (Byun and Ching, 1999; Byun and Schere, 2006) was used in its version 4.7.1 with the CB05 chemistry mechanism. Compared to its previous version, the model update includes

several new features (Foley et al., 2010), among them are gas phase chlorine chemistry, improved secondary organic aerosol (SOA) formation (Edney et al., 2007) and an updated representation of sea salt that considers reactions with nitric acid and the formation of coarse mode nitrate (Kelly et al., 2010) in the so-called AE5 aerosol mechanism. The model was run for an entire year with a spinup
165 time of 2 weeks. Standard profiles for the most important atmospheric pollutants were used as initial conditions. However, their effect on the simulated atmospheric concentrations of the substances in focus in this paper is negligible after the spinup.

The model was setup on a $72 \times 72 \text{ km}^2$ grid for entire Europe and subsequently on a nested $24 \times 24 \text{ km}^2$ grid for central Europe, see Fig. 4. The vertical model extent contains 30 layers up to 100
170 hPa in a sigma hybrid pressure coordinate system. 20 of these layers are below approx. 2km, the lowest layer extends to ca. 36m above ground. The evaluation area was restricted to the Greater North Sea region and some neighbouring section of the NE Atlantic, covering approximately half of the central European domain (see the red box in Fig. 4).

3.2 COSMO-CLM

175 The meteorological fields that drive the chemistry transport model were simulated with the COSMO-CLM mesoscale meteorological model (version 4.8) for the year 2008 (Geyer, 2014) using NCEP forcing data (Kalnay et al., 1996). This year was chosen because it does not contain very unusual meteorological conditions in Europe and can therefore be used to represent typical weather conditions in Europe. The same meteorological fields were used for the scenario runs, i. e. projected changes
180 due to climate change were not considered in order to avoid a mixture of effects, from emissions and meteorological data, in the resulting concentrations of air pollutants.

COSMO-CLM is the climate version of the regional scale meteorological community model COSMO (Rockel et al., 2008), originally developed by Deutscher Wetterdienst (DWD) (Steppeler et al., 2003; Schaettler et al., 2008). It has been run on a $0.22^\circ \times 0.22^\circ$ grid using 40 vertical layers up to 20 hPa
185 for entire Europe. COSMO-CLM uses the TERRA-ML land surface model (Schrodin and Heise, 2001), a TKE closure scheme for the planetary boundary layer (Doms and Schättler, 2002; Doms et al., 2011), cloud microphysics after Seifert and Beheng (2001, 2006), the Tiedtke scheme (Tiedtke, 1989) for cumulus clouds and a long wave radiation scheme following Ritter and Geleyn (1992). The meteorological fields were afterwards processed to match the CMAQ grid. As far as possible,
190 CMAQ uses the information that is provided by the meteorological input fields to calculate transport, transformation and loss of all gas phase and particulate species. The impact of the meteorological fields on the output of the chemistry transport model was investigated in detail in the articles by Matthias et al. (2009) and Bieser et al. (2011a).

3.3 Boundary conditions

- 195 Chemical boundary conditions for the outer model domain were taken from monthly means of the TM5 global chemistry transport model system (Huijnen et al., 2010) and were provided by the Dutch Royal Meteorological Institute (KNMI). The model results have been interpolated in time and space to provide daily boundary conditions for the 72 x 72km² CMAQ grid for Europe. Boundary conditions for the nested 24 x 24km² grid were calculated on hourly basis from the outer coarse grid.
- 200 They were kept the same for all scenarios in order to restrict the analysis of the effects of emission changes on shipping in North Western Europe.

3.4 Land based emissions

- The model runs were performed with full emissions from all relevant sources in the model domain. Land based emissions in hourly temporal resolution were produced with SMOKE EU (Bieser et al., 205 2011a) for the year 2011. They are based on officially reported EMEP emissions which are distributed in time and space using appropriate surrogates like population density, street maps or land use. Point sources were considered as far as information from the European point source emission register was available. The vertical distribution of the emissions was calculated online with the SMOKE model, the results are given by Bieser et al. (2011b). The land based emissions were kept 210 constant for all scenario model runs. Therefore the impact of reduced land based emissions, which can be expected for Europe in the year 2030, was not considered here. This was done to keep the the analysis clear and discuss only the effects of shipping instead of mixing it up with reductions of land based sources.

4 Impact of shipping on concentrations of pollutants

215 4.1 Situation today

- The results for today's air pollution due to shipping serve as a reference case for this study. They are discussed in detail in the accompanying paper by Aulinger et al. (2015). In brief it can be said that ships contribute significant amounts to the concentrations of NO₂, particle bound nitrate (NO₃⁻(p)) and particle bound sulphate (SO₄²⁻(p)). In summer, ozone is enhanced, too. High contributions from 220 shipping to the NO₂ and SO₂ concentrations are restricted to the open sea and the coastal areas in the southern North Sea and in Denmark (see the reference case in Fig. 5 and A.1). Nitrate and sulphate aerosol particles as well as ozone are secondary pollutants. They are transported far more inland but their relative contribution to concentrations at the coast is lower compared to NO₂ and SO₂.

There are large differences between summer and winter. **Partly, they can be ascribed to seasonal 225 differences in the emissions with higher shipping emissions in summer. Most of the differences in the concentrations are caused by atmospheric chemistry.** As a photochemical pollutant, ozone is only

increased during the summer months. The situation is similar for sulphate and nitrate aerosol. Both are formed via oxidation path ways that include the photochemically formed OH radical. Therefore, the conversion rate of SO_2 into $\text{SO}_4^{2-}(\text{p})$ and of NO_2 into $\text{NO}_3^-(\text{p})$ in summer is higher than in winter. This leads to higher contributions of shipping emissions to the concentrations of these aerosol components in summer. On the other hand, total nitrate aerosol concentrations are much lower in summer compared to winter, because the gas-to-particle partitioning between HNO_3 and $\text{NO}_3^-(\text{p})$ is temperature dependent with higher particulate nitrate concentrations at low temperatures.

4.2 Scenarios for the North Sea in 2030

- 235 To derive the contribution of ships to the selected pollutant concentrations two model runs, one including and one excluding shipping emissions, were performed. The difference is regarded as the contribution of ships to the individual pollutant. For the scenarios the difference between two model runs with different shipping emissions is regarded as the change in the contribution of ships in the respective scenario.
- 240 We mainly discuss the consequences of changes in the NO_x emissions from ships because here we see the main differences between the scenarios (Fig. 2). Additionally, the strict rules for SO_2 came into force in the North Sea ECA on 1 January 2015, and there are only small differences between the scenarios with respect to SO_2 emissions in the North Sea (Fig. 3). This will be further discussed in section 4.2.4.
- 245 NO_x emissions from ships have an impact on the NO_2 concentrations, on nitrate aerosol and on ozone. It can be expected that NO_2 concentrations increase due to ship emissions. The impact of NO_x emissions from ships on ozone will be different between winter and summer. While in summer increased NO_x emissions will lead to increased ozone under most weather conditions and in most regions, this will have almost no effect in winter.
- 250 In the following, maps illustrating changes in the contribution of shipping on NO_2 , nitrate aerosol and ozone concentrations in 2030 will be shown for the scenarios No ECA, ECA SCR 16 (Tier III in 2016), ECA SCR 21 (Tier III in 2021) and ECA opt. The color coded relative changes refer to the impact of the shipping emissions on the concentrations of selected pollutants. The latter is given in reference maps which are results of the reference model run considering shipping emissions
- 255 from 2011. The changes in the other scenarios will be illustrated in time series for different North Sea coastal regions (see Figure 4), each of them comprising about 9200 km^2 . These time series nicely show the highly variable impact of shipping emissions, which depends to a large extent on the weather conditions and the concentrations of pollutants from other sources.

4.2.1 Nitrogen dioxide

- 260 Scenario No ECA is reflecting a steady increase in shipping activity disregarding the implementation of stricter rules for NO_x emissions. This will lead to an increase in the contribution of shipping to the

average NO₂ concentrations by more than 30 % in large areas of the North Sea region (Fig. 5b). The largest increase can be seen in the English Channel and the south western North Sea while Norway and Sweden exhibit lower increase rates of around 20%.

265 Figure 5c displays scenario ECA SCR 21 in which the Tier III rules for new ships come into force in 2021. This means that in 2030 a large part of the fleet will still follow the less strict Tier I and Tier II regulations. Very few ships older than 30 years will not have to comply with any of the NO_x regulations. As a consequence, the contribution of ships to average NO₂ concentrations will be higher than in the case with Tier III regulations from 2016 onwards (Fig. 5d). In large parts of the North
270 Sea the contribution of shipping to NO₂ concentrations will be higher than today.

Scenario ECA SCR 16 on average still shows a moderate increase in the NO₂ concentrations caused by ships compared to the situation today (Fig. 5d). While in the English Channel and the southern North Sea the concentrations decrease by a few percent only, they decrease by more than 10% in the north western parts of the North Sea and in particular at the British, Norwegian and Swedish coast.

275 This is caused by the fact that the traffic to the main North Sea ports in Rotterdam, Hamburg and Antwerp will still increase and ships will become bigger, resulting in a rather small concentration decrease in the English Channel. Today, smaller and older ships travel to the smaller harbours in the North Sea area. However, many of them will be replaced after 2016, which means that a large fraction of those ships will comply with Tier III. This will lead to a reduction of the contribution of
280 shipping emissions to NO₂ concentrations in the central and northern part of the North Sea.

The contribution of shipping to NO₂ concentrations will be drastically reduced in the case of scenario ECA opt when all ships comply with Tier III rules for NO_x emissions (Fig. 5e). The simulations show a reduction of approximately 80% all over the North Sea region compared to today.

All reductions in the contribution of shipping to NO₂ concentrations have a similar magnitude and
285 regional distribution in winter and summer. On average the impact of shipping is slightly higher in winter compared to summer with a larger increase in scenario No ECA and ECA SCR 21 and a smaller decrease in scenario ECA SCR 16 and ECA opt. All maps for the winter case can be seen in the appendix (Fig. A.3).

Figure 6 shows a time series of the contribution of shipping to the daily average NO₂ concentrations
290 at the coasts of Belgium and the Netherlands (see Fig. 4 for the region). Of the scenarios for 2030, scenario ECA SCR 16 shows a slight decrease in the contribution of shipping to the NO₂ concentrations compared to today. If Tier III will be implemented in 2021 (scenario ECA SCR 21) or not at all until 2030 (scenario No ECA), the contribution of shipping to the NO₂ concentration will be higher than today. Large reductions of NO₂ from shipping, on some days more than 4 µg/m³,
295 are only achieved when all ships and not only new-buildings follow the Tier III regulations. Time series for the other region are included in the appendix (Fig. A.6).

4.2.2 Nitrate Aerosol

Nitrate aerosol (NO_3^- (p)) is formed in the atmosphere as a consequence of the oxidation of NO_2 .

The amount of aerosol particles formed highly depends on the presence of other pollutants, in par-

300 ticular on the availability of ammonia (NH_3). Ammonia mainly stems from agricultural activities.

The regions with the highest ammonia emissions are western France, the Benelux countries, western

Germany and Denmark. Particulate ammonium nitrate preferentially exists in winter, at low tem-

peratures. At higher temperatures ammonium nitrate particles decompose into gaseous ammonia

and nitric acid. Therefore, nitrate aerosol concentrations all over Europe are much lower in summer

305 compared to winter. On the other hand, oxidation of NO_2 is much more effective in summer leading

to a higher contribution of shipping to nitrate aerosol compared to winter.

In summer, the emission scenarios show very similar results for nitrate aerosol and for NO_2 (see

Fig. 7). In scenario No ECA the contribution of shipping to nitrate aerosol concentrations increases

310 by more than 30 % over sea and by 25 % or more in large areas of central Europe and in southern

Scandinavia (Fig. 7b). In scenario ECA SCR 21 (Figure 7c) large areas of the North Sea, and in

particular northern France, show an increase in nitrate aerosol from shipping while in other areas the

situation will remain unchanged. Scenario ECA SCR 16 (Fig. 7d) shows a decrease in the contri-

bution of shipping to nitrate aerosol concentrations by 7-10% in the north eastern part of the North

Sea while in the south western part a small increase by 5 - 10% can be observed. Again, in scenario

315 ECA opt the contribution of shipping to nitrate aerosol will be reduced by 60 - 80%.

In winter, nitrate aerosol concentrations are only marginally affected by shipping emissions. For this

reason the results of the scenario runs do not show reliable patterns of changes in NO_3^- (p) concen-

trations caused by changing shipping emissions when given as relative changes. Therefore, they are

not shown.

320 4.2.3 Ozone

NO_x emissions from ships have a strong influence on the atmospheric ozone concentrations. Ozone

is formed out of NO_2 and atmospheric oxygen in the presence of sunlight. Volatile organic com-

pounds (VOC) help to transform emitted NO into NO_2 , thereby enhancing the ozone formation

significantly. On the other hand NO destroys ozone, leading to low ozone concentrations during

325 night-time when no photolysis of NO_2 takes place. This leads to a strong diurnal cycle of the ozone

concentration and a large difference between winter and summer levels with much higher ozone

concentrations in the summer. Furthermore, increased NO_x emissions may cause additional ozone

formation in presence of sufficiently high VOC concentrations. If the VOC levels are comparably

low, more NO_x causes ozone destruction.

330 Here, we look at the impact of shipping emissions on the daily mean ozone values. Figure 8 shows

maps of the distribution of changes in the contribution of ships to mean ozone concentration for the

different scenarios. Fig. 8a shows that shipping causes about $7 \mu\text{g}/\text{m}^3$ additional ozone (summer average value) in large parts of the North Sea and in Denmark. On the other hand, there is only a small increase in ozone in the English Channel, where NO_x concentrations are high. The effect of ozone destruction by additional NO_x emissions under low VOC conditions can be clearly seen in scenario No ECA (Fig. 8b). Reductions in ozone concentrations caused by shipping emissions, by partly more than 80%, are clearly noticeable in the English channel, the south western North Sea and the surrounding coast line. On the other hand, the shipping contribution to increased ozone concentrations will be enhanced by more than 20% far from the main shipping areas in Central Europe, Ireland and the Northern UK.

Scenario ECA SCR 21 (Fig. 8c) contains higher NO_x emissions, leading to decreased ozone in the English channel and higher values in Central Europe, France, Ireland and the UK. Scenario ECA SCR 16 (Fig. 8d) shows a lower shipping contribution to ozone concentrations in the North East of the North Sea region and an almost unchanged situation in the South West. In the case of ECA opt, ozone caused by shipping emissions is significantly reduced by 40-60% all over the modeling domain, except for the English channel where a significant increase is found.

Fig. 9 displays a time series of the daily average ozone concentrations in northern Germany. It can be seen that the ozone values would be lower in summer, on some days more than $10 \mu\text{g}/\text{m}^3$, if ships emitted as little NO_x as in the ECA opt scenario. On the other hand they would be slightly higher in winter.

An analysis of the different regions reveals that the days with concentrations higher than $120 \mu\text{g}/\text{m}^3$ (a value recommended by the World Health Organization, WHO) would decrease significantly by 50% or more without shipping emissions in all regions except the Netherlands (see Table 1). The scenarios for 2030 do not show big differences in the number of days with concentrations above $120 \mu\text{g}/\text{m}^3$. While small increases in the number of days can be expected if Tier III rules were not implemented (scenario No ECA), the only case with a strong decrease in exceedance days is scenario ECA opt.

4.2.4 Sulphur dioxide and sulphate aerosol

Sulphur dioxide (SO_2) emissions from ships are directly related to the sulphur content of ship fuels. In the scenarios for 2030 all ships will follow the same rules for sulphur, which allow 0.1% S in the fuel in the North and Baltic Sea ECA and 0.5% outside of it. Therefore, the scenarios do not differ much in terms of sulphur emissions. The main difference is between the ECA SCR and ECA LNG scenarios, because LNG does not contain any sulphur at all which makes the sulphur emissions from these ships even lower than for all other ships that comply with the 0.1% S rule inside the North Sea area.

In Fig. 10 changes in the contribution from shipping to the SO_2 and SO_4 concentrations for the scenario ECA SCR 16 and ECA LNG 16 are shown for summer. In the ECA LNG 16 scenario

sulphur dioxide and sulphate aerosol concentrations are even further reduced than in the ECA SCR 16 scenario. The reductions are between 40% and 60% for the SCR case and between 60% and 80% 370 for the LNG case. The reductions are slightly higher for SO₂ compared to sulphate, however the reductions for sulphate are more widespread than those for SO₂. The results for the other scenarios are very close to those in Fig. 10 which is why they are not shown here. More maps are included in the appendix (Fig. A.1, A.2, A.4, A.5).

4.2.5 PM_{2.5}

375 Particulate matter with a diameter less than 2.5 μm (PM_{2.5}) originating from shipping emissions is mainly formed through a conversion of gaseous SO₂ and NO₂ into particulate nitrate (NO₃⁻(p)) and sulphate (SO₄²⁻(p)). The amount of these secondary aerosol components depends critically on the level of NH₃ emissions, which are a prerequisite for the formation of ammonium sulphate and ammonium nitrate in the atmosphere. The area where the highest contribution of shipping to the 380 PM_{2.5} concentrations is noticeable is south east of the main shipping lanes, in North West France, Belgium, The Netherlands and North Germany. These are areas with high ammonia emissions from agricultural activities.

Reductions in the contribution of shipping to PM_{2.5} are visible in all scenarios (see Fig. 11). This is caused by the significant effect the sulphur reductions in the ship fuel has on sulphate aerosol 385 concentrations. In scenario ECA opt the PM_{2.5} reduction is the largest. Here, also nitrate aerosol is significantly reduced.

The time series for PM_{2.5} concentrations and the respective reductions in the different scenarios can be seen for Northern Germany in Fig. 12. All scenarios except for No ECA show reductions in PM_{2.5} on almost all days. Again, the largest reductions can be seen for ECA opt. There is no clear 390 seasonal trend for the reductions although total PM_{2.5} is higher in winter compared to summer.

5 Conclusions

This paper investigates the effects of different future developments of shipping emissions in the North Sea area on air quality in the North Sea region. The main differences between the scenarios for 2030 concern nitrogen oxide emissions. They could be significantly lowered by using exhaust cleaning techniques or alternative fuels like LNG. Additionally, international regulations for a 395 mandatory reduction of nitrogen oxide emissions in the North and Baltic Sea areas are under debate in the International Maritime Organization.

It was found that the expected increase in ship traffic in the North Sea will lead to enhanced levels of NO₂, nitrate aerosol and ozone in large areas of North West France, Belgium, The Netherlands, 400 North Germany and Denmark if no emission reduction measures will be taken. For secondary pollutants like ozone and aerosols, this effect is more pronounced in summer compared to winter. In

winter shipping does not contribute much to elevated levels of aerosol concentrations.

The effect of emission reduction measures depends on the year of implementation. If already in 2016 new ships needed to follow the new Tier III rules for new-buildings, the concentrations of

405 NO₂, nitrate aerosol and ozone in 2030 would be on the same level as today. This means that the emission reductions of the new ships are compensated by an increased ship traffic. If it took another five years until Tier III regulations were in place, the concentrations of NO₂, nitrate aerosol and ozone which are caused by ship traffic would be higher in 2030 compared to today. The difference to the implementation of the Tier III rules in 2016 would be about 10-15%. Significant reductions

410 can only be achieved if all ships, not only new-buildings, followed strict NO_x emission limitations.

The situation is different for sulphur dioxide, sulphate aerosol particles and also for PM_{2.5}. Regulations on lower sulphur levels in ship fuels have been implemented on 1 January 2015 for the North Sea and will be in place for all seas in 2020 (or the latest in 2025). This will significantly reduce the impact of shipping on SO₂ and sulphate aerosol concentrations. As a consequence of lower sulphate

415 aerosol concentrations PM_{2.5} concentrations will also be reduced. The use of LNG as alternative fuel would further reduce sulphur emissions and therefore also SO₂ and PM_{2.5} concentrations.

Our model study shows that all effects of shipping emissions on air quality differ largely by region and season, depending on the pollutant in focus. Gaseous primary pollutants like NO₂ and SO₂ have a short life time. Consequently, their effects can mainly be seen close to the shipping lanes.

420 Aerosols, which are formed through oxidation in the atmosphere can be transported over large distances. Contributions of shipping to nitrate, sulphate and PM2.5 concentrations can be seen far inland. For ozone, future emission reductions of NO_x could even lead to enhanced concentrations in regions that already today have high NO_x and low VOC concentrations like in the English Channel.

However, this will depend on the future development of other NO_x emission sources, too. These

425 were not taken into account here, as has been done for climate change neither, in order to focus on shipping effects and facilitate the interpretation of emission changes in this sector. Because it can be expected that in particular land based anthropogenic NO_x emissions in Europe will further decrease until 2030, the relative contribution of shipping emissions to NO₂ and nitrate pollution levels in the North Sea area will be higher than demonstrated here. Ozone might increase in regions where it was

430 found to decrease in the scenarios No ECA and ECA SCR 16, namely in the English Channel. When this region, where ozone formation is currently VOC limited, turns into a NO_x limited region, NO_x emissions from shipping will enhance ozone concentrations in the entire study area.

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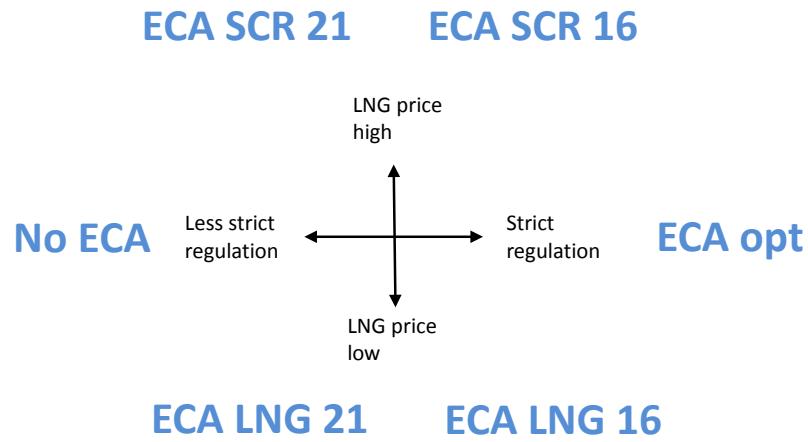
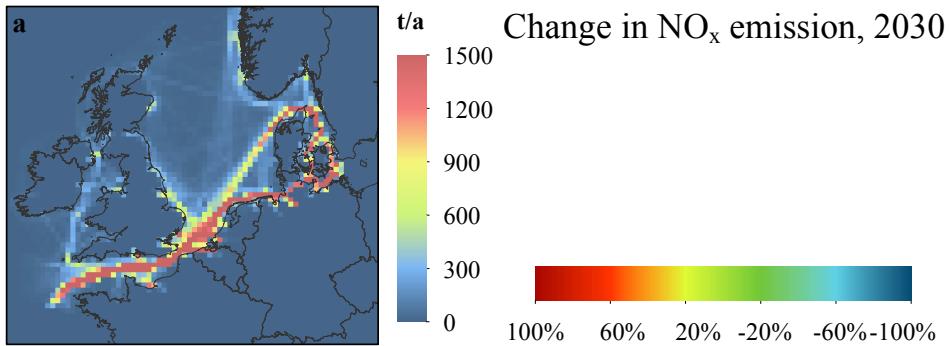
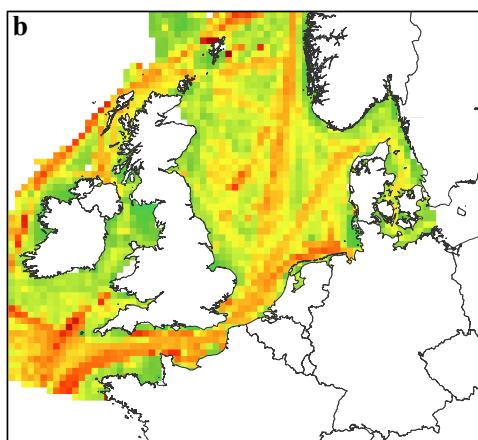


Fig. 1. Scenario identifiers for technical developments and legislation with respect to ship emissions for 2030.

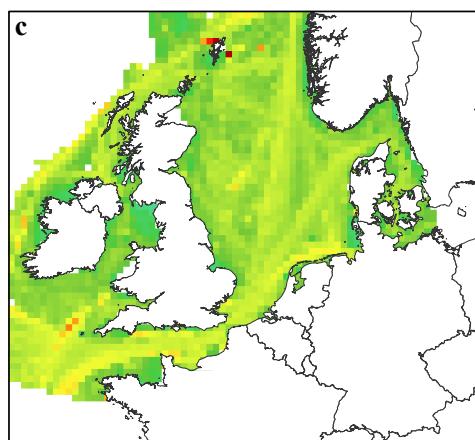
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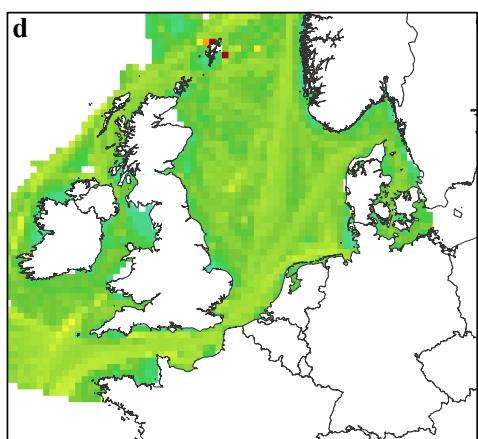
No ECA



ECA SCR 21



ECA SCR 16



ECA opt

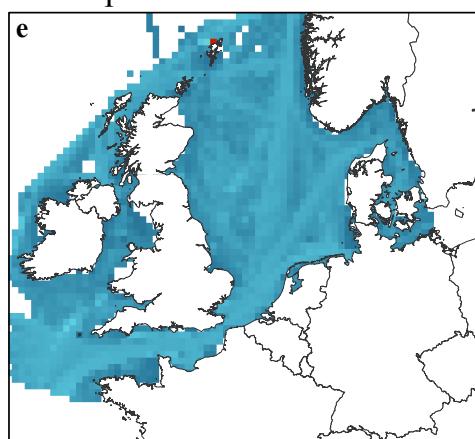


Fig. 2. NO_x emissions from ships, a) annual totals in t per grid cell of 24 x 24 km². Emission changes for the scenarios b) No ECA, c) ECA LNG, d) ECA SCR, and e) ECA opt for 2030. **No values are shown in grid boxes where the NO_x emissions from ships were below 0.5 t/a per grid cell of 24 x 24 km².**

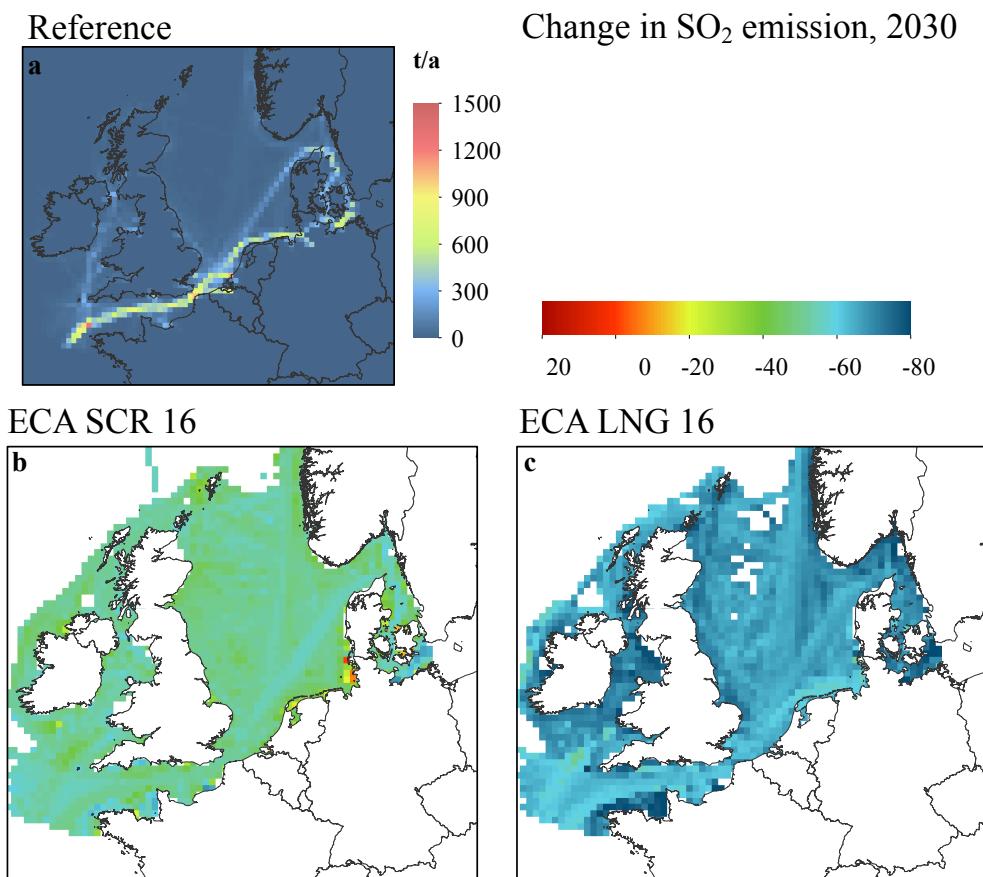


Fig. 3. SO₂ emissions from ships, a) annual totals in t per grid cell of 24 x 24 km². Emission changes for the scenarios b) ECA SCR 16, and c) ECA LNG 16 for 2030. No values are shown in grid boxes where the SO₂ emissions from ships were below 0.5 t/a per grid cell of 24 x 24 km².

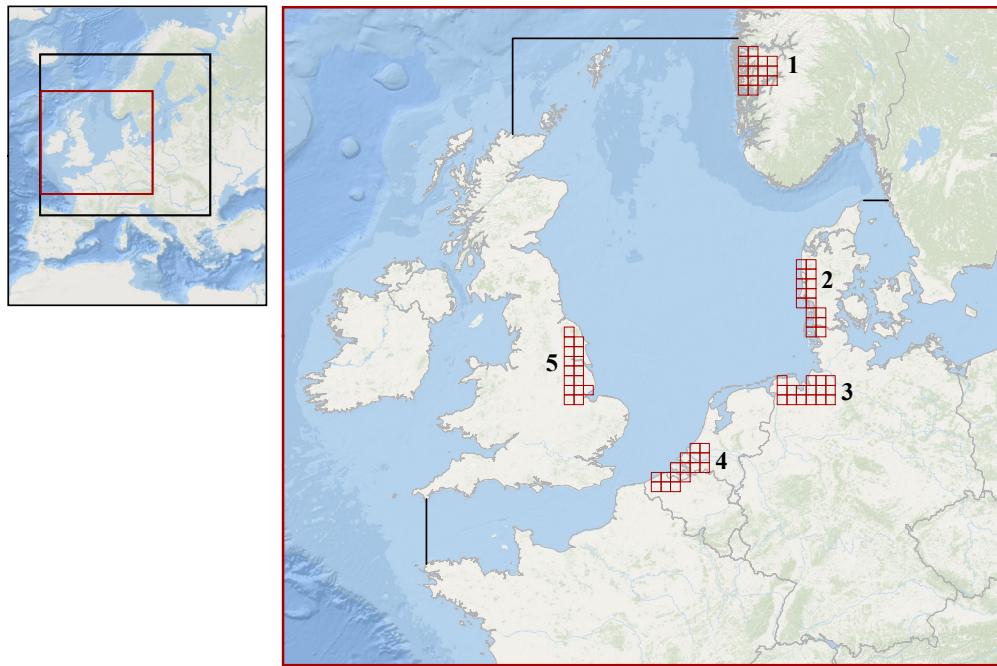


Fig. 4. Left: Modelling domains, outer domain with $72 \times 72 \text{ km}^2$ resolution (outer black line), inner domain with $24 \times 24 \text{ km}^2$ resolution (inner black line) and the evaluation area (red line). Right: Evaluation area including the greater North Sea region illustrating also the five regions (namely 1 to 5) for which time series of pollutant concentrations have been derived from CMAQ modelling results.

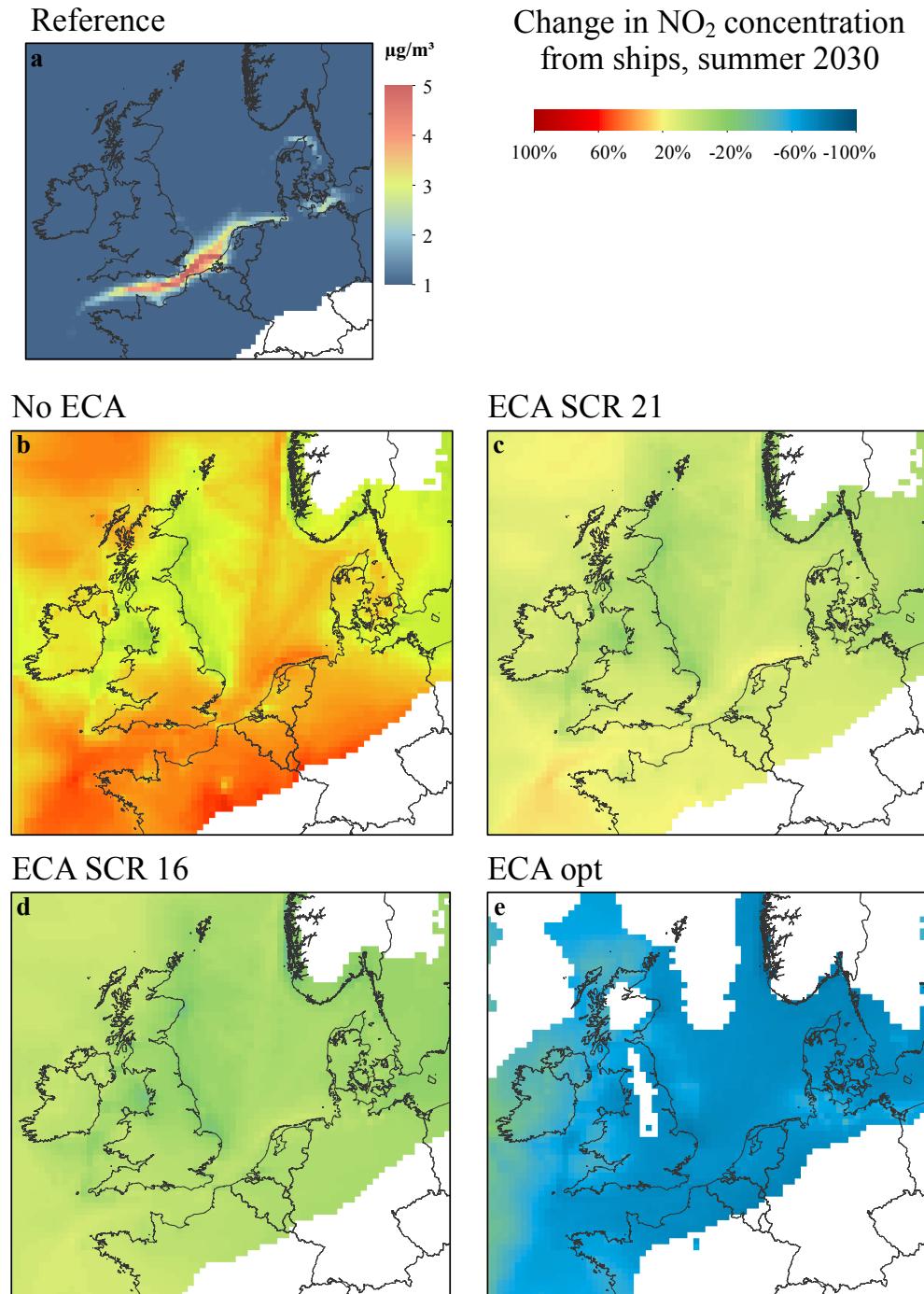


Fig. 5. Contribution of shipping to the total NO₂ concentrations in summer (JJA) (a) today (Reference) and change in the scenarios (b) No ECA, (c) ECA SCR 21, (d) ECA SCR 16, (e) ECA opt. No values are shown in grid boxes where the contribution from shipping to the NO₂ concentrations in either the reference or the scenario case was below 0.05 $\mu\text{g}/\text{m}^3$.

NO2NL

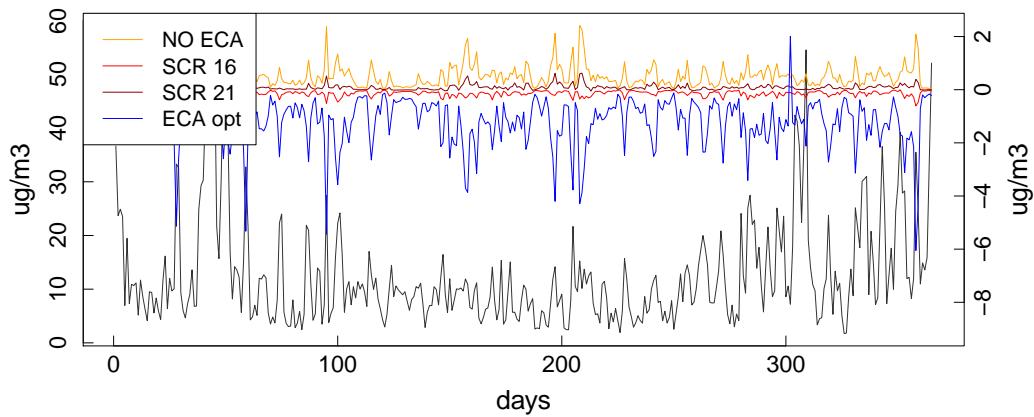


Fig. 6. Time series of daily average NO_2 concentrations in $\mu\text{g}/\text{m}^3$ (black, left y-axes) and the contribution of shipping to the NO_2 concentrations in the coastal areas of Belgium and the Netherlands (region 4) for all scenarios (colored, right y-axes).

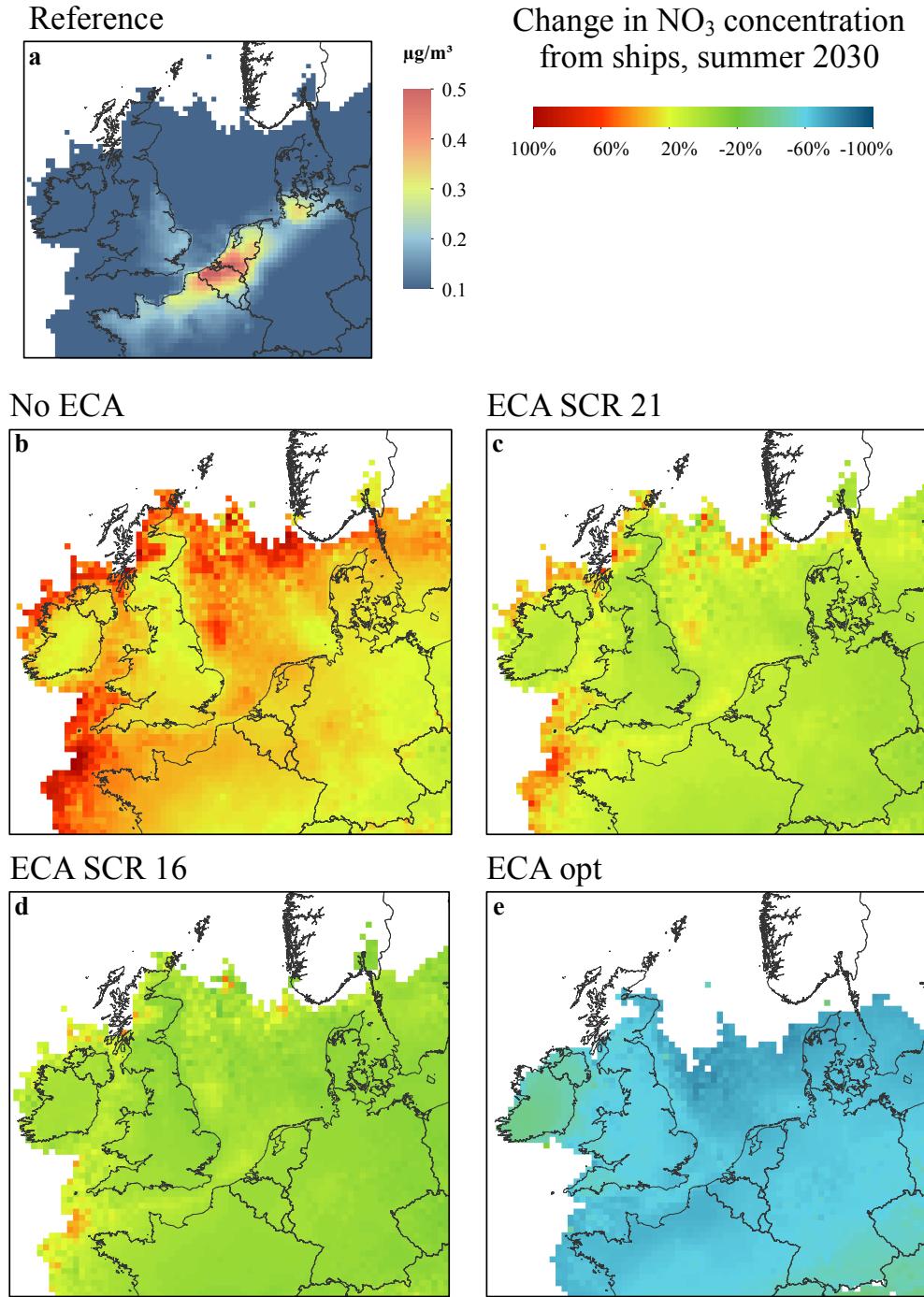


Fig. 7. Contribution of shipping to the total NO_3^- concentrations in summer (JJA) (a) today (Reference) and change in the scenarios (b) No ECA, (c) ECA SCR 21, (d) ECA SCR 16, (e) ECA opt. No values are shown in grid boxes where the contribution from shipping to the NO_3^- concentrations in either the reference or the scenario case was below $0.005 \mu\text{g}/\text{m}^3$.

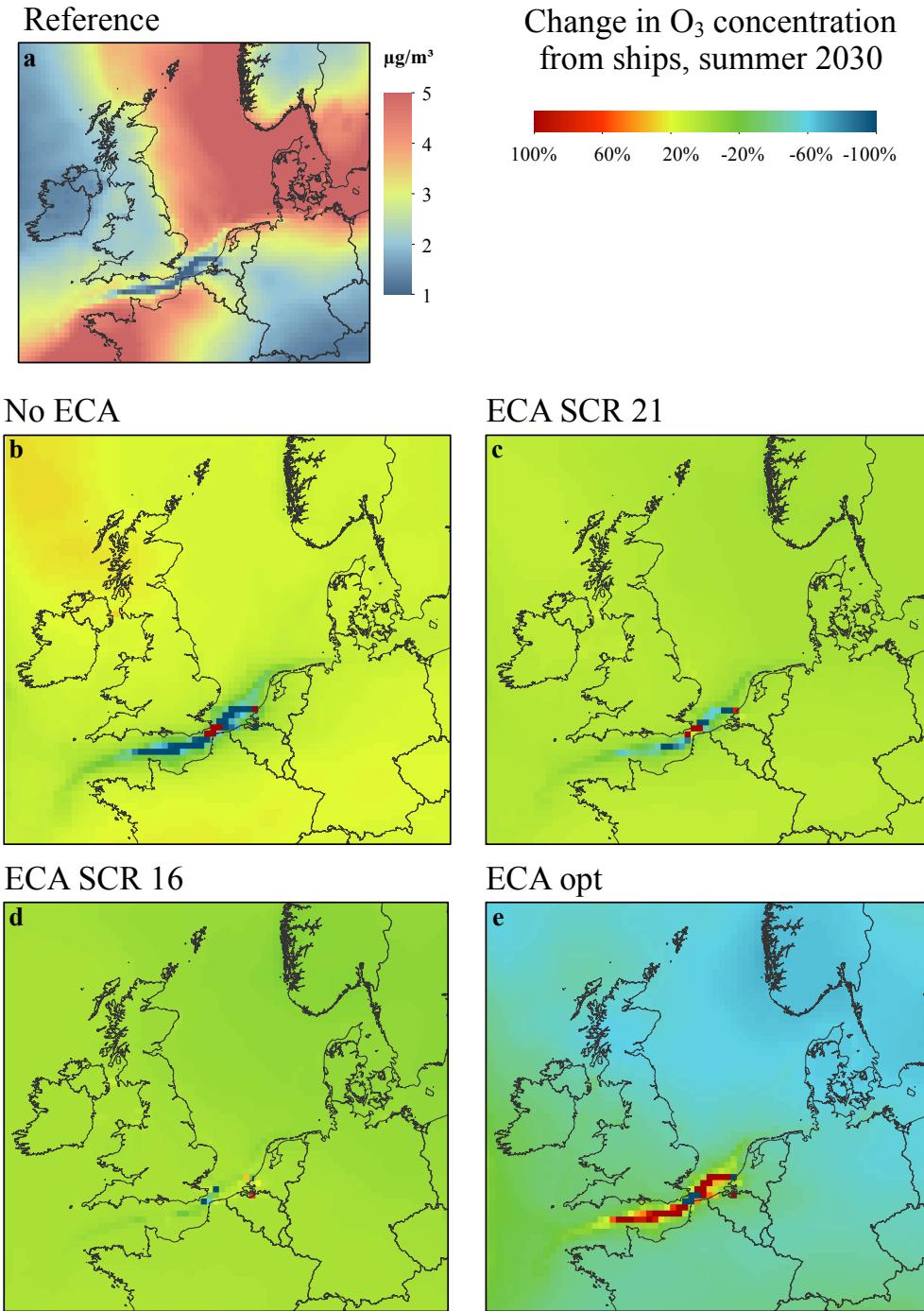


Fig. 8. Contribution of shipping to the mean O₃ concentrations in summer (JJA) (a) today (Reference) and change in the scenarios (b) No ECA, (c) ECA SCR 21, (d) ECA SCR 16, (e) ECA opt.

O3DE

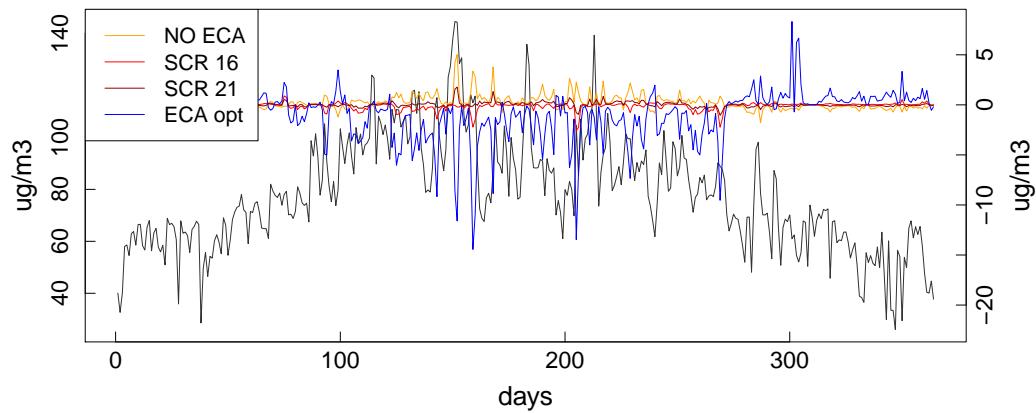


Fig. 9. Time series of daily average ozone concentrations in $\mu\text{g}/\text{m}^3$ (shaded grey, left y-axes) and the contribution of shipping to the ozone concentrations in the coastal areas of Germany (region 3) for all scenarios (right y-axes). "noship" denotes the changes if all ship emissions were cut to zero

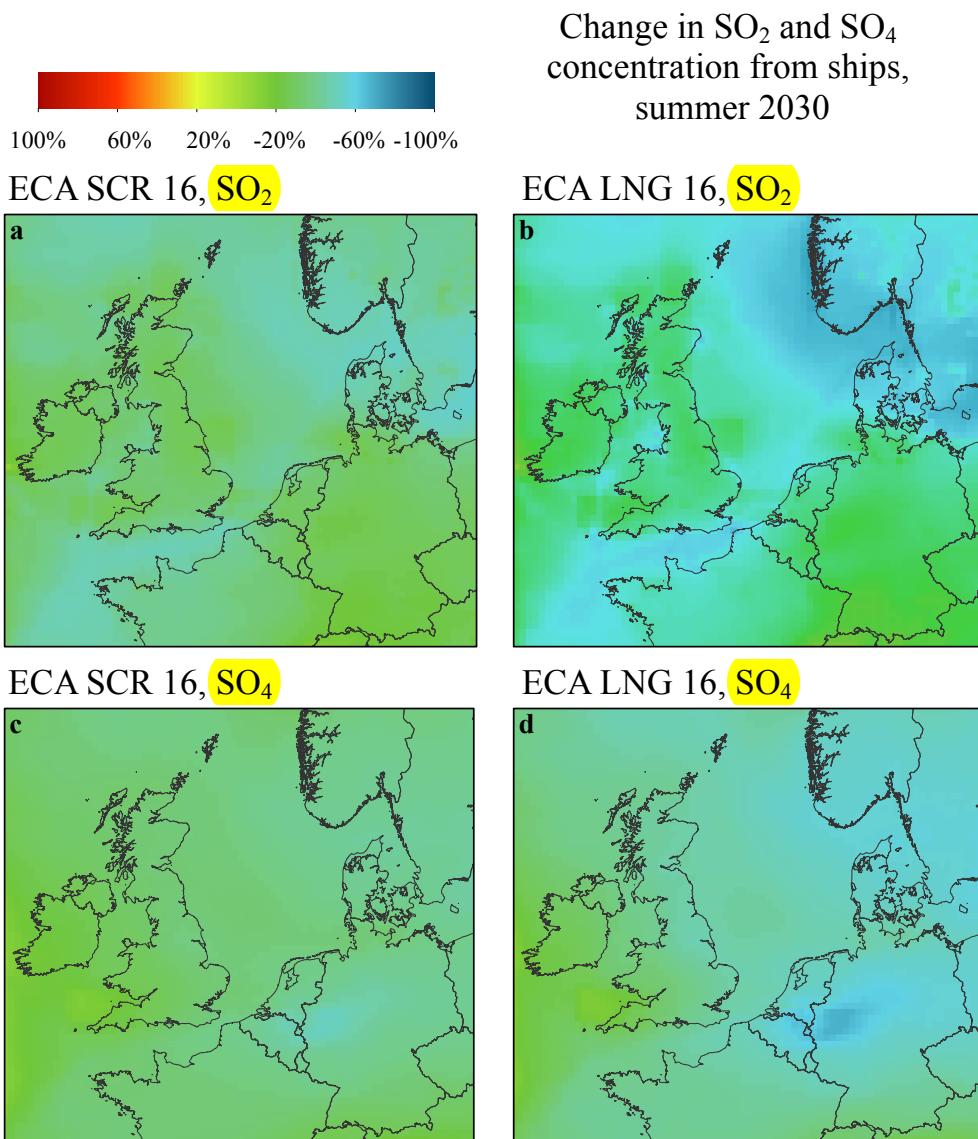


Fig. 10. Change in the contribution of shipping to the total (a and b) SO₂ and (c and d) SO₄ concentrations in summer (JJA) for the scenarios ECA SCR 16 (left), ECA LNG 16 (right), in relation to the reference case. See Figures A.1 and A.2 for the reference concentrations

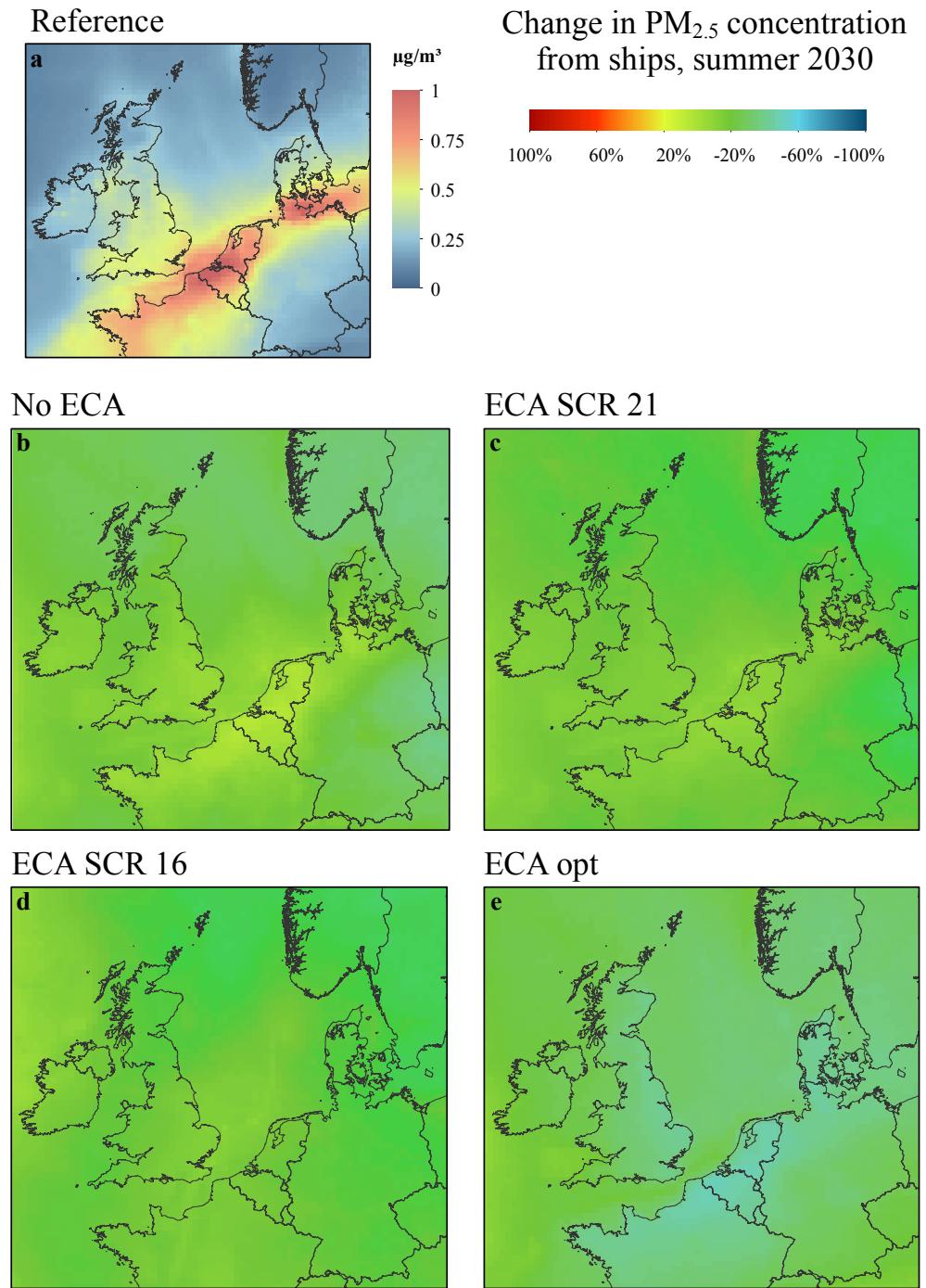


Fig. 11. Contribution of shipping to the total PM_{2.5} concentrations in summer (JJA) (a) today (Reference) and change in the scenarios (b) No ECA, (c) ECA SCR 21, (d) ECA SCR 16, (e) ECA opt.

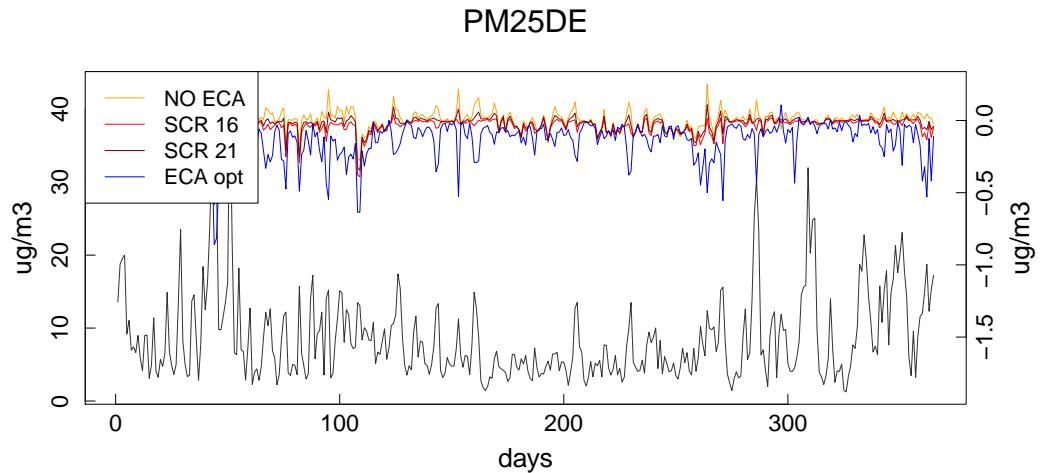


Fig. 12. Time series of daily average PM2.5 concentrations in $\mu\text{g}/\text{m}^3$ (black, left y-axis) and the contribution of shipping to the PM2.5 concentrations in the coastal areas of Germany (region 3) for the main scenarios (colored, right y-axis).

Table 1. Number of days with 8-hour maximum ozone concentrations greater than $120 \mu\text{g}/\text{m}^3$ in 2030 in selected regions around the North Sea for the scenarios No ECA, ECA SCR 16, ECA SCR 21, and ECA opt.

	1	2	3	4	5
2011	9	19	27	46	29
No ECA	9	20	29	46	29
ECA SCR 16	8	18	26	46	29
ECA SCR 21	8	18	27	46	29
ECA opt	6	12	16	45	22

Appendix A

545 Supplementary information on other seasons and other pollutants

A1 Sulphur containing species in summer

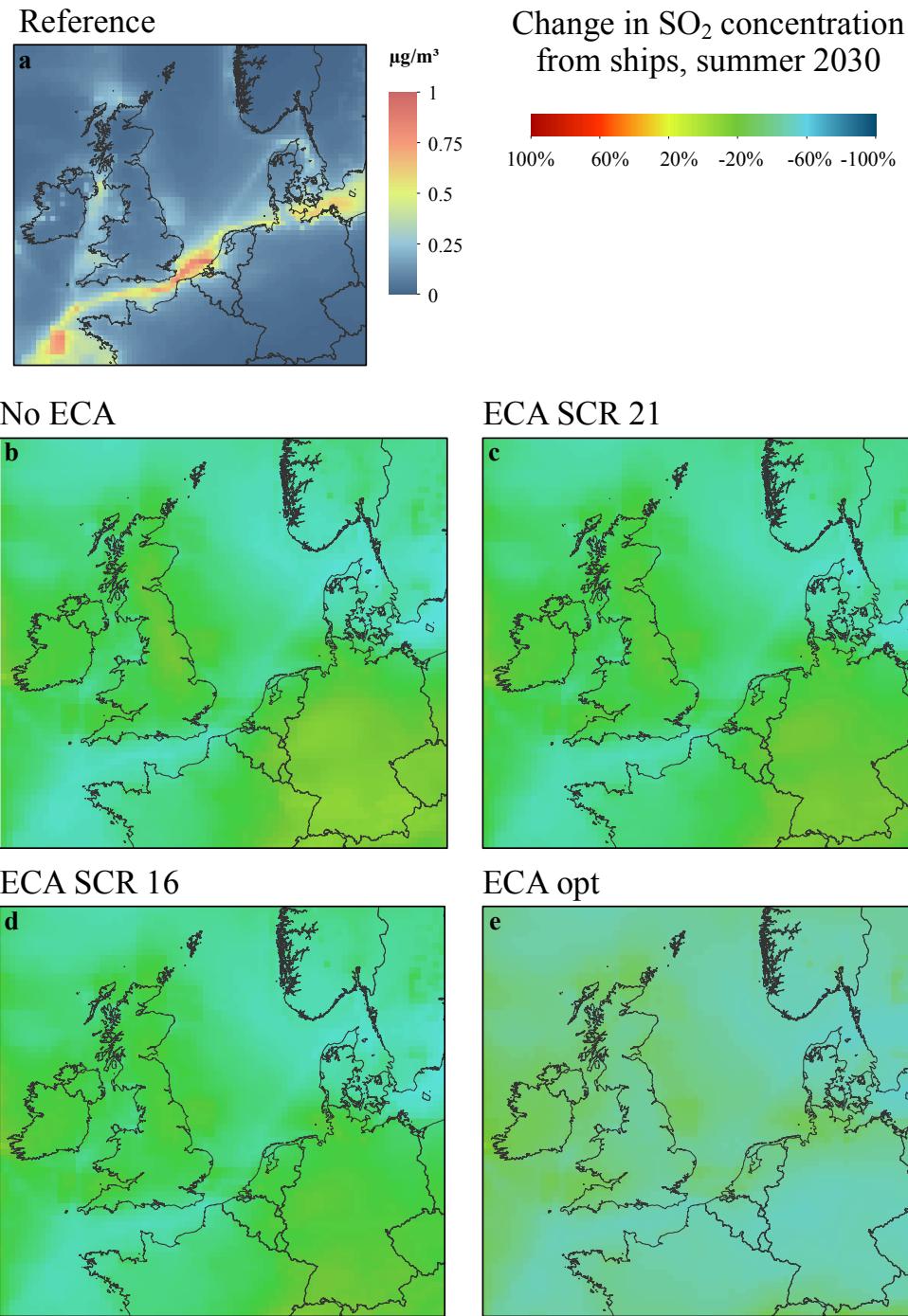


Fig. A.1. Change in the contribution of shipping to the total SO_2 concentrations in summer (JJA) compared to the reference case (a) for the scenarios (b) No ECA, (c) ECA SCR 16, (d) ECA SCR 21, and (e) ECA opt.

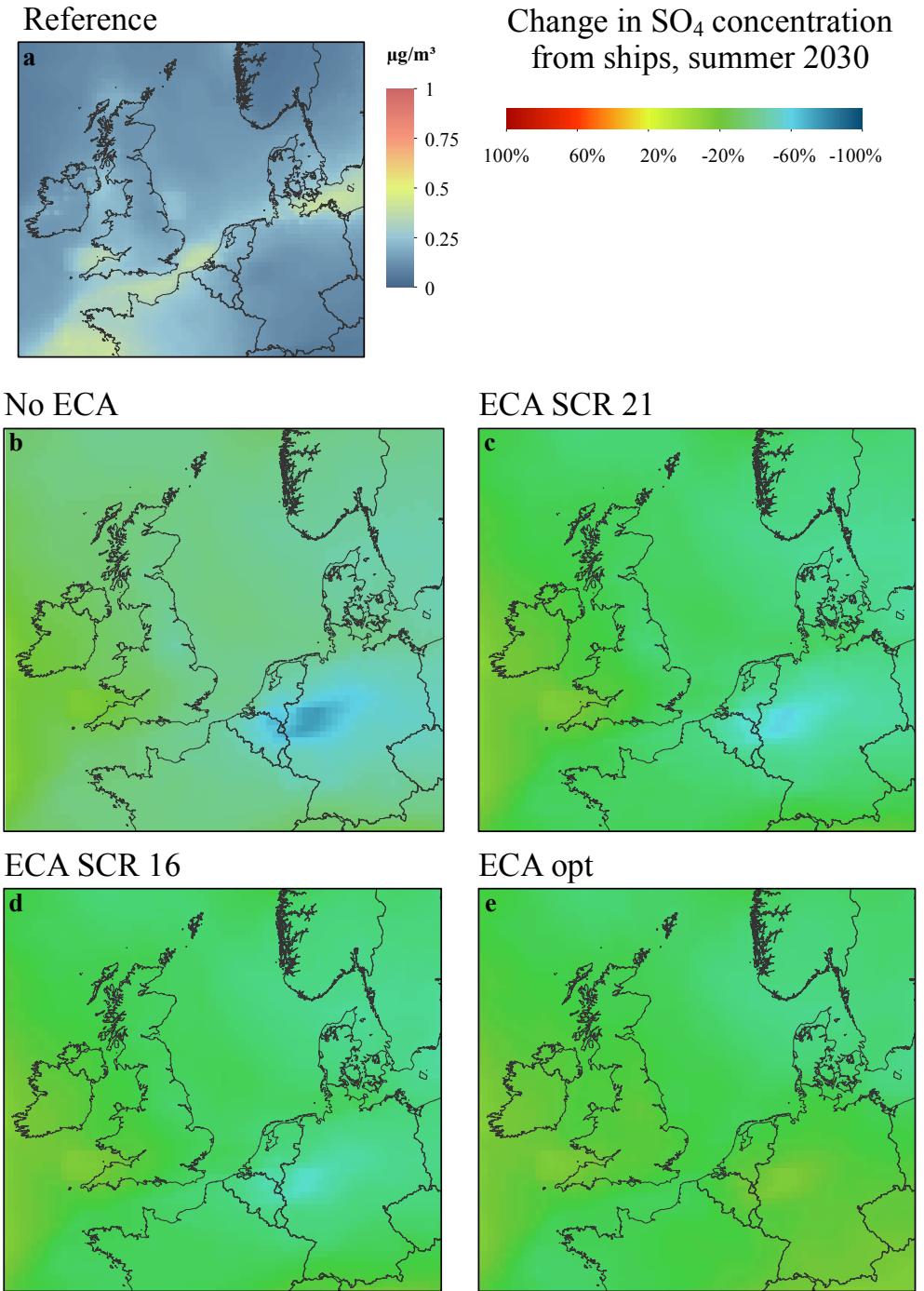


Fig. A.2. Change in the contribution of shipping to the total SO₄ concentrations in summer (JJA) compared to the reference case (a) for the scenarios (b) No ECA, (c) ECA SCR 16, (d) ECA SCR 21, and (e) ECA opt.

A2 Contribution of shipping to air pollution in winter

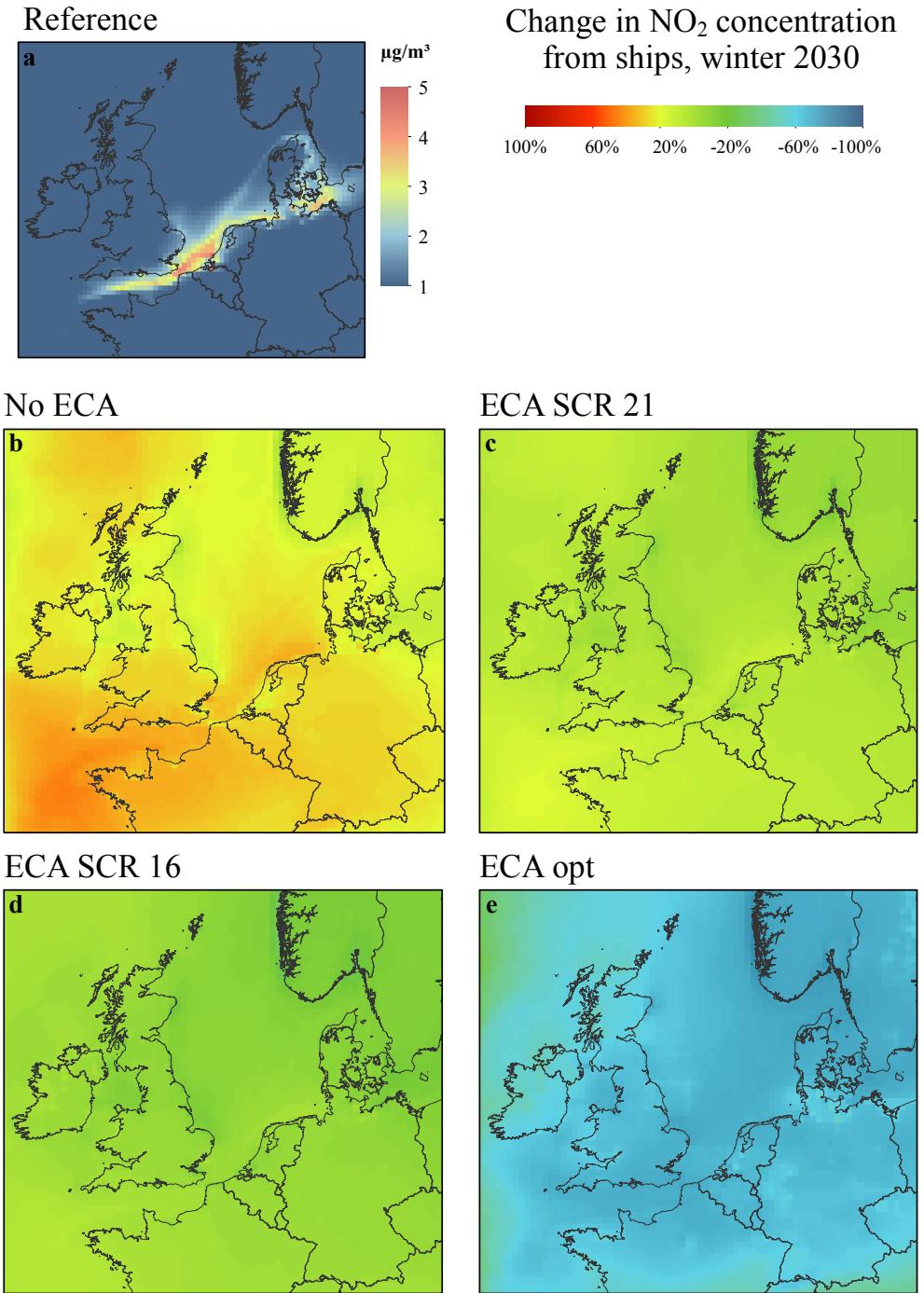


Fig. A.3. Change in the contribution of shipping to the total NO₂ concentrations in winter (DJF) compared to the reference case (a) for the scenarios (b) No ECA, (c) ECA SCR 16, (d) ECA SCR 21, and (e) ECA opt.

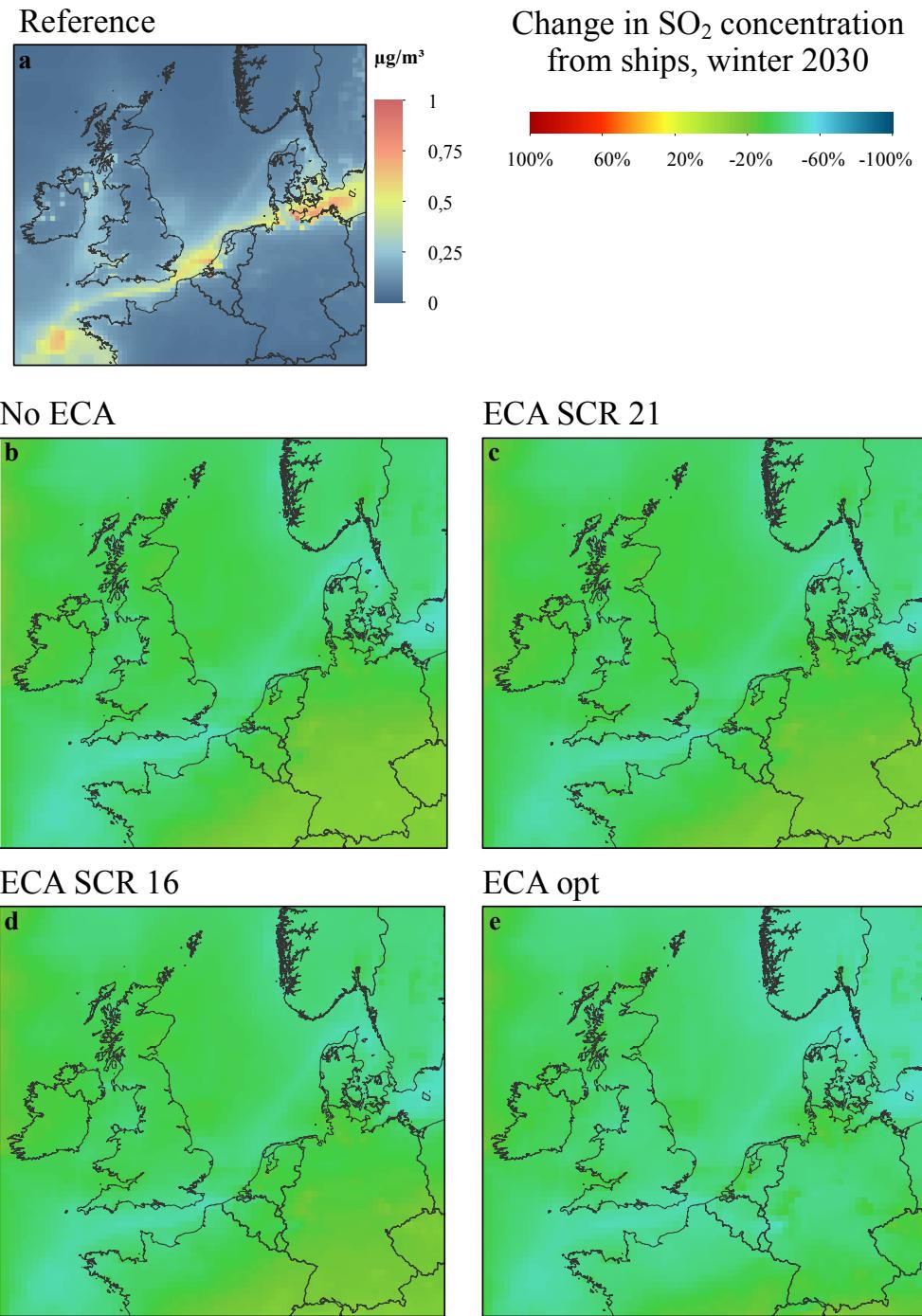


Fig. A.4. Change in the contribution of shipping to the total SO₂ concentrations in winter (DJF) compared to the reference case (a) for the scenarios (b) No ECA, (c) ECA SCR 16, (d) ECA SCR 21, and (e) ECA opt.

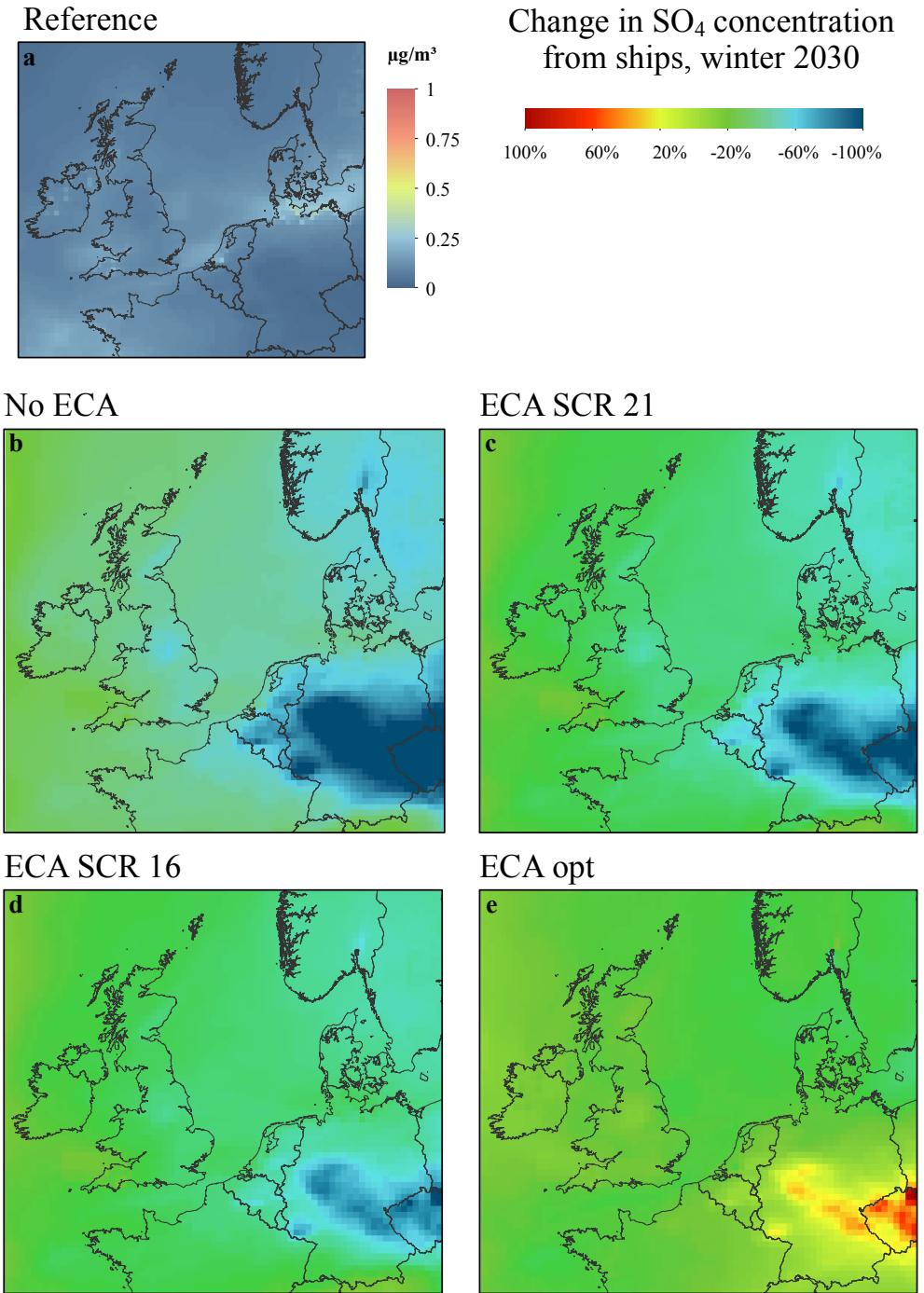


Fig. A.5. Change in the contribution of shipping to the total SO₄ concentrations in winter (DJF) compared to the reference case (a) for the scenarios (b) No ECA, (c) ECA SCR 16, (d) ECA SCR 21, and (e) ECA opt.

A3 Time series in selected areas

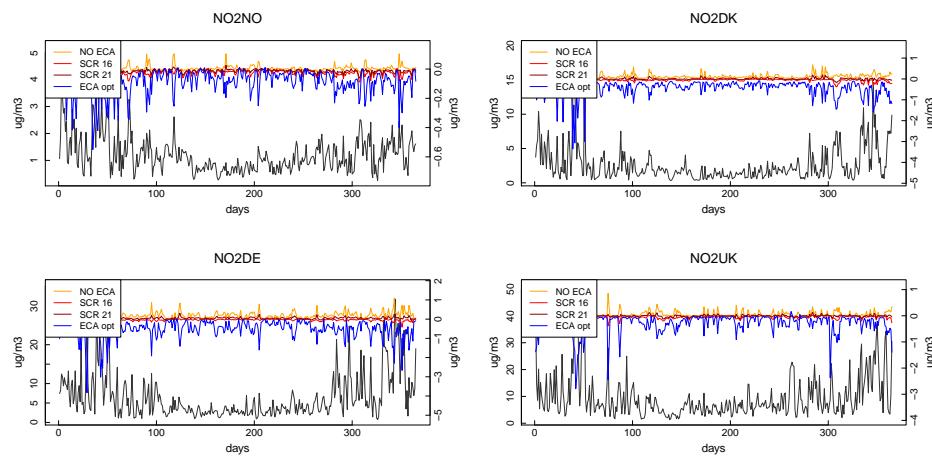


Fig. A.6. Time series of daily average NO_2 concentrations in $\mu\text{g}/\text{m}^3$ (black, left y-axes) and the contribution of shipping to the NO_2 concentrations in the coastal areas of Norway (region 1), Denmark (region 2), Germany (region 3), and Great Britain (region 5) for all scenarios (right y-axes).

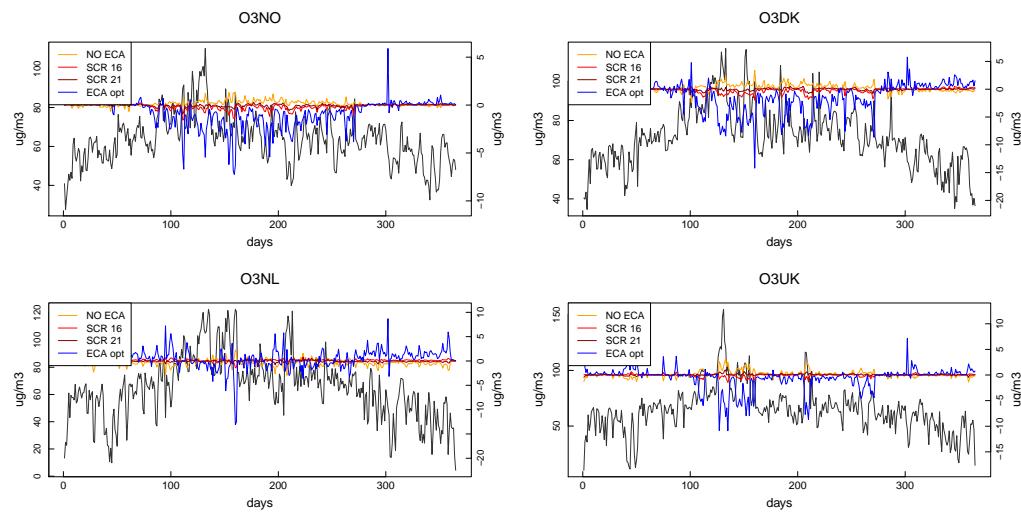


Fig. A.7. Time series of daily average O_3 concentrations in $\mu\text{g}/\text{m}^3$ (black, left y-axes) and the contribution of shipping to the O_3 concentrations in the coastal areas of Norway (region 1), Denmark (region 2), Belgium and The Netherlands (region 4), and Great Britain (region 5) for all scenarios (right y-axes).

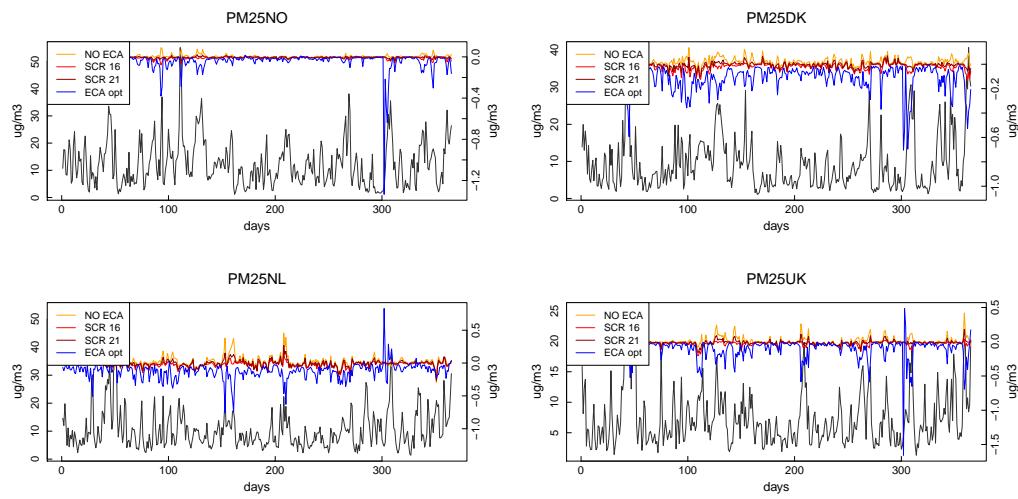


Fig. A.8. Time series of daily average PM_{2.5} concentrations in $\mu\text{g}/\text{m}^3$ (black, left y-axes) and the contribution of shipping to the PM_{2.5} concentrations in the coastal areas of Norway (region 1), Denmark (region 2), Belgium and The Netherlands (region 4), and Great Britain (region 5) for all scenarios (right y-axes).