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THE CONTRIBUTION OF SHIP EMISSIONS TO AIR POLLUTION IN THE NORTH SEA REGIONS

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ABSTRACT

As a consequence of the global distribution of manufacturing sites and the increasing international division of labour, ship traffic is steadily increasing and is becoming more and more important as an origin of air pollution.

This study investigates the impact of ship emissions in coastal areas of the North Sea under conditions of the year 2000 by means of a regional chemistry transport model which runs on a sufficiently high resolution to study air pollution in coastal regions. It was found that northern Germany and Denmark in summer suffer from more than 50 % higher sulphate, nitrate and ammonium aerosol concentrations due to contributions from ships. The implementation of a sulphur emission control area (SECA) in the North Sea, as it was implemented at the end of 2007, directly results in reduced sulphur dioxide and sulphate aerosol concentrations while nitrate aerosol concentrations are slightly increased.

Ship emissions lead to significantly enhanced air pollution by secondary inorganic aerosols in North Sea coastal areas.

Key words: ship emissions, aerosol pollution, regional air quality modelling, coastal air quality

1. INTRODUCTION

Ship emissions can have considerable impact on atmospheric concentrations of several important pollutants especially in coastal areas (Tsyro and Berge, 1997, Lawrence and Crutzen, 1999, Endresen et al., 2003). The most important ones are CO_2, NO_x, SO_2, CO, hydrocarbons, and primary and secondary particulates because of their role as e.g. greenhouse gas (CO_2), their contribution to acid rain (NO_x, SO_2), and/or their impact on human health (CO, hydrocarbons, particulates) (Lloyd´s Register Engineering Services, 1995, Corbett and Fischbek, 1997, Corbett et al., 1999). Corbett et al. (2007) have

recently shown that ship emissions lead to an increase in PM2.5 ambient air concentration and therewith are responsible for an increase of premature deaths due to cardiopulmonary diseases and lung cancer. The future use of low sulphur fuels could enhance air quality and possibly prevent large health impacts due to ship emissions (Winebrake et al., 2009).

The southern North Sea with the large harbours in Rotterdam and Hamburg is one of the regions with the highest ship traffic in the world. This study focuses on the impact of NO_x and SO_2 emitted by ships on the pollution distribution in North Sea coastal areas. Their contribution to elevated air concentrations of sulphate, nitrate and ammonium aerosol particles is investigated. To what extent a sulphur emission control area, which is implemented in the North Sea since the end of 2007, also leads to a significant reduction of particle concentrations is subsequently investigated. In previous studies about the impact of ship emissions on air quality in Europe global models with coarse horizontal resolutions were used (Dalsøren et al., 2007, Collins et al., 2009), which makes it rather difficult to estimate the effects of ship emissions on coastal areas and on the population living there. A coarse grid resolution might additionally raise concerns about non-linear chemical effects, particularly for ozone, as stated in the only study about the effect of ship emissions that was run on a moderate grid resolution of 50 x 50 km^2 (Jonson et al., 2009).

For the estimation of the ship emissions in this study a bottom-up approach on the basis of ship movement data together with average engine loads and emission factors available in literature (Cooper and Gustafsson, 2004) is used to generate a ship emission

inventory that is of sufficient spatial resolution to investigate coastal gradients in aerosol concentrations. This inventory serves as input for the Models-3 Community Multiscale Air Quality modelling system (CMAQ) (Byun and Ching, 1999), a state-of-the-art Eulerian chemistry transport model.

2. METHODS

2.1 Model system

CMAQ version 4.5 is set up on a 54 x 54 km^2 grid for Europe and on a 18 x 18 km^2 grid for the greater North Sea area. It uses the CBM IV chemical mechanism (Gery et al., 1998) with aerosol and aqueous chemistry extensions. The aerosol is represented by three log-normal size modes (Binkowski and Roselle, 2003) and it includes secondary inorganic aerosols as sulphate, ammonium and nitrate, sea salt, organic carbon and elemental carbon.

The model is driven by meteorological fields that were derived from MM5 (Grell et al., 1995) model runs that were driven by ERA40 6 hourly global reanalysis data on a 1° x 1° grid. We used four dimensional data assimilation of the ERA40 fields in MM5 and applied the more sophisticated physical parameterization schemes like Reisner 2 (Reisner et al., 1998) for cloud microphysics, Kain Fritsch 2 (Kain, 2004) for cumulus representation and the MRF scheme (Hong and Pan, 1996) for the boundary layer to produce meteorological data that is as close as possible to wind, temperature and

humidity observations. The quality of the meteorological data has been tested and the results are described in a separate paper (Matthias et al., 2009).

The boundary conditions for the CMAQ simulations were taken from MOZART (Horowitz et al., 2003, Niemeier et al., 2006) model results for the year 2000. The data has a resolution of 1 x 1° and one day. It includes the most important gas phase species (e.g. O_3, CO, NO_x, SO_4, OH, HNO_3) but no aerosol particles. The concentrations were interpolated to the boundary of the outer CMAQ domain. It is one grid cell thick and is updated hourly.

Land based emission data for the nitrogen, sulphur and volatile organic compounds as well as for aerosol particles was provided by IER Stuttgart based on EMEP area emissions and EPER point source emissions (Friedrich and Reis, 2004). The distribution of the data to the CMAQ grids was based on geostatistical data like population density and street maps. The temporal variation of the emissions follows typical activity profiles for the different SNAP sectors.

Matthias et al. (2008) have shown that the model is applicable to North Sea coastal regions by comparing model results to measured air concentrations and depositions. It has also been shown that the model system gives reliable results for secondary inorganic aerosol particle concentrations (Matthias, 2008) that are in focus of this paper.

2.2 Ship emissions

Several authors have estimated global ship emissions with different approaches in recent years (e.g. Whall et al., 2002, Eyring et al., 2005, Endresen et al., 2007), however these global data sets are on a grid with rather coarse resolution and they do not reflect the interannual variability of the ship emissions. The assessment of air pollution in coastal regions and their possible effects on the population in these regions requires a more detailed ship emission inventory. We used a bottom-up approach that is based on individual ship movements, information about the ship characteristics and emission factors for NO_x and SO_2.

The vessel database was purchased from Lloyds Marine Intelligent Unit (LMIU). It consists of a vessel characteristic database and a vessel movement database and it includes all commercial vessels equal to or greater than 100 gross tonnages (GT). The vessel characteristic database comprises information on e.g. vessel type, engine type, numbers of engines, engine speed at the crankshaft, fuel type, power and maximum speed. The ship movement database provides information on ship movements for the year 2000 in parts of Europe (riparian states of the North and Baltic Sea, Atlantic coast of France, Spain and Portugal). This includes 15625 ships which perform 651825 movements on 58324 different routes. The ship movement database consists of previous departure, arrival, departure and next arrival places and dates with a daily time resolution.

Emission factors (power-based in g/kWh) used in this study are obtained from Cooper and Gustafsson (2004) who compiled these emission factors for Sweden´s international reporting duties. The emissions from ships are affected by several characteristics of the

ships as well as the fuel used (see Whall et al. (2002) for details). The engine type determines the combustion conditions and therefore the emissions of some pollutants.

The procedure how to obtain the ship emission dataset is displayed in Figure 1. The vessel movement database provides the basis for calculating the routes of the vessels. It is assumed that all vessels take the shortest routes between two ports at sea. For the required temporal resolution of the ship movements it is assumed that all vessels arrive and depart at 6 a.m. in they travel for more than one day. If they leave the same day they leave the port at 6 p.m. or even earlier if they call another port the same day. The travelling time is distributed equally to all passed grid boxes on their route. The ship emissions per grid box and time step are calculated for the different pollutants by means of the corresponding emission factors and the engine power of the particular vessel.

Figure 2 shows the land based and the additional ship emissions for NO_x and SO_2 in the inner 18 x 18 km² grid for January and July 2000. It can be seen that the relative contribution of the ship emissions, particularly for SO_2, is much larger in July than in January because the land based emissions related to heating are much lower in summer. Land based sulphur emissions were significantly decreased in the last two decades because wood and coal combustion has decreased, flue gas desulphurization is implemented in large power plants and cars drive with low sulphur gasoline. On the other hand the sulphur content in bunker oil is still high and the ship traffic in the North Sea area has increased in recent years.

2.3 Model runs

The land-based and marine emission datasets serve as input into the Eulerian air quality modelling system CMAQ which computes the concentration and deposition distribution over Europe. To investigate the contribution of ship emissions to air pollution in coastal areas three different model runs of the complete year 2000 were performed with CMAQ, one model run including all land-based emissions and the ship emissions, a reference model run just including the land-based emissions and a model run where the sulphur emissions in the North and Baltic Seas were reduced by 45 %. This corresponds to the case that all ships in the SECA use fuels that contain only 1.5 % sulphur instead of 2.7 %, which is the typical sulphur content of residual oils that are mainly used by large ocean going vessels.

The seasonal impact is considered by comparing January and July monthly average concentrations as representatives for winter and summer. We then investigated the effects of NO_x and SO_2 emissions on NO_2 and SO_2 concentrations as well as nitrate, sulphate and ammonium aerosol concentrations in the greater North Sea area.

3. RESULTS

First the results for the case that ships use standard bunker oil and that the emission factors given by Cooper and Gustafsson (2004) can be applied as they are given is presented. Then the differences that arise from a reduction of 45 % in the sulphur content of these fuels is discussed.

3.1 Nitrogen dioxide and sulphur dioxide

The results of the CMAQ model runs for NO_2 and SO_2 are displayed in Figures 4 and 5. Both species show higher concentrations in winter than in summer (left panels in Fig 4 and Fig. 5). This is mainly caused by the lower mixing heights in winter and therefore the suppressed vertical distribution of the pollutants that are emitted close to ground. For SO2, the lower land based emissions in summer amplify the differences between summer and winter.

Both NO_2 and SO_2 concentrations are significantly enhanced by the ship emissions (middle panels of Fig. 4 and Fig. 5). In the regions of the main ship traffic, NO_2 concentrations are typically increased by 5 ppb, while the increase in SO_2 concentrations is about 1-2 ppb. The concentrations over water may be increased by 50 – 100 % in winter and by more than 500 % in summer. In Skagerrak and Kattegat these values might be even higher. However, these large effects are limited to the open sea where NO_2 and SO_2 concentrations would be low without ship traffic and only small coastal areas reveal heavily increased pollution by NO_2 and SO_2.

3.2 Nitrate aerosol

Nitrate aerosol concentrations are significantly higher in winter than in summer (see Fig. 6). Again this is partly caused by less efficient vertical mixing in winter but ambient temperatures are another important factor that determines the nitrate aerosol concentrations. Nitrate aerosol is in thermodynamic equilibrium with gaseous nitric acid. At low temperatures particulate nitrate dominates while at higher temperatures the

equilibrium is shifted towards nitric acid. Particulate nitrate exists in the as ammonium nitrate aerosols. These can only be formed if gaseous ammonia is available, and this largely depends on the sulphate concentrations. Atmospheric ammonia is first used to neutralize sulphate and form ammonium sulphate aerosol. Particulate nitrate can only exist if excess ammonia would be available.

Additional nitrate due to NO_x ship emissions is mainly formed over land and there is more additional nitrate in summer than in winter. Since the aerosol formation needs free ammonia, this only happens over land where ammonia is emitted and the concentrations are high enough. The amount of nitrate caused by NO_x emissions from ships is lower in winter because oxidation of NO_2 into HNO_3 is much slower in winter compared to summer. This leads to almost unchanged nitrate concentration in winter but in summer nitrate aerosol concentrations might be increased by more than 50 % all over Denmark, large parts of northern Germany, the Netherlands, Belgium and France (right side of Fig. 6).

3.3 Sulphate aerosol

The concentration differences between summer and winter are less pronounced for sulphate aerosol particles. Generally the levels are comparable with somewhat higher aerosol concentrations in summer. As for NO_2, SO_2 is oxidized more efficiently in summer leading to higher sulphate concentrations in summer. In contrast to nitrate, sulphate aerosol can also be built in regions where no gaseous ammonia exists. This is

because in presence of sulphate ammonium nitrate aerosol particles may break up and gaseous nitric acid and ammonium sulphate are formed (Ansari and Pandis, 1998). Ship emissions contribute much more to the total SO_2 emissions in the modelling domain in summer, therefore the increase in sulphate aerosol concentrations in summer is significantly larger than in winter. In winter sulphate aerosol concentrations might be enhanced by about 10 % in Denmark, south Sweden and south Norway but in summer these areas suffer from more than 50 % enhanced sulphate aerosol concentration which are, in contrast to nitrate, already on a high level during this season. The German North Sea coastline is affected in a similar way. Unlike nitrate, sulphate aerosols are also formed over water and particularly the English Channel shows a large increase in sulphate aerosols caused by SO_2 emissions from ships.

3.4 Ammonium aerosol

Ammonium aerosol particles exist in close connection with sulphate and nitrate aerosols. Like in most other chemistry transport model systems, in CMAQ ammonium sulphate aerosol is formed as long as ammonia is present and until all available sulphate is consumed to form particles. If there is still ammonia left, i.e. the molar ratio of ammonia to sulphate is larger than 2, additional ammonium nitrate is formed.

The high ammonium concentrations in winter are in accordance with the higher nitrate values in winter and in summer ammonium is lower because of the higher temperatures that prevent the formation of nitrate. Similar to sulphate and nitrate the largest increase

in ammonium concentrations due to ship emissions is modelled for July and mainly Denmark and northern Germany are affected. Here, the ammonium concentrations are increased by about 50 %.

3.5 Annual cycle

The annual cycle of the NO_x and SO_2 emissions in the complete model domain, both with and without ship emissions, are displayed in Fig. 9 a). While the NO_x emissions are more or less constant over the year, SO_2 emissions are slightly lower in summer compared to winter. The reason for the differences is that coal and wood burning in households is much lower in summer. For SO_2 the domain averaged emissions are increased by 19 % in winter and 30 % in summer due to ships, for NO_x the values range between 18 % in winter and 22 % in summer.

As already seen in the previous paragraphs, this has almost no effect on the aerosol concentrations in winter, the domain averaged increase of the aerosol mass is around 5 % only. In summer, on the other hand the domain averaged aerosol concentrations are at maximum 22 % (for ammonium in July), 27 % (for nitrate in July) and 29 % (for sulphate in July), respectively, higher than in the reference case. As seen in sections 3.2 to 3.4 the aerosol concentrations may be increased by more than 50 % in regions 100 – 200 km downwind the coast.

3.6 Effect of a sulphur emission control area in the North Sea

290 Since 22 November 2007 ships travelling in the North Sea east of 4°W (east of 5 °W in
291 the region of the English channel) and south of 62°N are not allowed to use fuels with a
292 sulphur content of more than 1.5 % of the mass. Such a SECA was already implemented
293 in the Baltic Sea in May 2006 (IMO, 2008). Typical sulphur contents in the most used
294 residual oils are about 2.7 %. This means that the new SECA results in a reduction of
295 the sulphur emissions by ships in this area by about 45 %.
296
297 This has been taken into account in a third model run that has been performed for the
298 year 2000. Ship based sulphur emissions in the North and the Baltic Seas were reduced
299 by 45 % in both the 54 x 54 km^2 grid run and the nested run on the 18 x 18 km^2 grid that
300 covers the North Sea and its coastlines. The relative increase of nitrate, sulphate and
301 ammonium aerosol particles in January and July are shown in Fig. 10. The results
302 should be compared to the right columns of Figs 6, 7 and 8. The effects on the aerosol
303 concentration in winter are further reduced or remain more or less unchanged. As
304 intended, the sulphate concentrations are significantly reduced, a reduction is also
305 visible for ammonium. Nitrate is only slightly increased, although one might have
306 expected from the interplay between sulphate, nitrate and ammonium that significantly
307 more nitrate can be formed if sulphate is reduced (see e.g. Lauer et al., 2009).
308
309 An average over the whole model domain shows that the additional SO_2 from ship
310 emissions is reduced by 42 %, additional sulphate aerosols decrease by 38 % and
311 additional ammonium aerosols by 20 %. Nitrate aerosols were 7 % higher compared to
312 the standard run with sulphur rich ship fuels.
313

4. CONCLUSIONS

Ship emissions account for a large part of the secondary inorganic aerosol formation in North Sea coastal areas. In northern Germany, Denmark and southern Sweden the sulphate aerosol concentration might be increased by 50 % or more caused by the burning of sulphur rich bunker oil in ship engines. Increased nitrate and ammonium aerosol concentrations by more than 50 % in summer affect mainly Denmark and northern Germany. The largest increase in the ammonium and nitrate concentrations are seen over land, where gaseous ammonia is emitted which favours the formation of ammonium nitrate aerosol. NO_2 and SO_2 concentrations are mainly increased along the shipping routes at sea and directly at the coastline while the relative influence of the ship emissions on the NO_2 and SO_2 concentration levels at land is rather low. These distinctions between air pollution over land, that is dominated by aerosols and over sea, that is dominated by direct gaseous emissions from ships are visible thanks to the high spatial resolution of the model runs.

According to our model results, the introduction of sulphur emission control areas in the Baltic Sea in 2006 and the North Sea in 2007 will lead to significantly reduced SO_2 and sulphate aerosol concentrations in the greater North Sea area. Also ammonium aerosol particle mass will decrease by about 20 % while nitrate aerosol mass on the other hand will increase. This is related to the fact that more nitrate can be formed if sulphate is reduced and ammonia is still available to form ammonium nitrate. However, the effect is relatively small with an increase of the nitrate concentrations by 7 %, averaged over

the year and the whole inner model domain. This means that the implementation of a sulphur emission control area in the North Sea effectively reduces aerosol concentrations and that the reduction in sulphate aerosol mass is not counterbalanced by increased nitrate aerosol.

Nevertheless, ship emissions contribute to a large extent, particularly in summer, to air pollution in coastal areas. It can be expected that the secondary aerosol formation over the North Sea will have additional adverse effects on the eutrophication of North Sea coastal waters and ecosystems at the eastern border of the North Sea. Besides a further reduction of sulphur emissions from ships that is already foreseen in the newest IMO regulations, emission control measures to reduce NOx emissions from ships will certainly be needed to further reduce particle formation in coastal areas and the input of nitrogen and sulphur into sensitive ecosystems like the Wadden Sea.

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Figure captions

Figure 1: Outline of the procedure to generate the ship emission dataset

Figure 2: Average land based nitrogen dioxide emissions for January and July 2000 (left side) and the respective ship emissions (right side).

Figure 3: Average land based sulphur dioxide emissions for January and July 2000 (a) and c)) and the respective ship emissions (b) and d)).

Figure 4: Average nitrogen dioxide concentration for January and July 2000 (a) and d)), the absolute increase caused by ship emissions (b) and e)) and the relative increase caused by ship emissions (c) and f)).

Figure 5: Average sulphur dioxide concentration for January and July 2000 (a) and d)), the absolute increase caused by ship emissions (b) and e)) and the relative increase caused by ship emissions (c) and f)).

Figure 6: Average nitrate aerosol concentration for January and July 2000 (a) and d)), the absolute increase caused by ship emissions (b) and e)) and the relative increase caused by ship emissions (c) and f)).

Figure 7: Average sulphate aerosol concentration for January and July 2000 (left side), the absolute increase caused by ship emissions (middle) and the relative increase caused by ship emissions (right side).

Figure 8: Average ammonium aerosol concentration for January and July 2000 (a) and d)), the absolute increase caused by ship emissions (b) and e)) and the relative increase caused by ship emissions (c) and f)).

Figure 9: Annual cycle of NO_x and SO_2 emissions (a)) and aerosol concentrations for sulphate, nitrate and ammonium (b)), both averaged over the complete model domain.

Figure 10: Relative increase of nitrate (a) and d)), sulphate (b) and e)) and ammonium (c) and f)) concentrations caused by ship emissions in January and July 2000 considering 45 % reduced sulphur content in ship fuels.

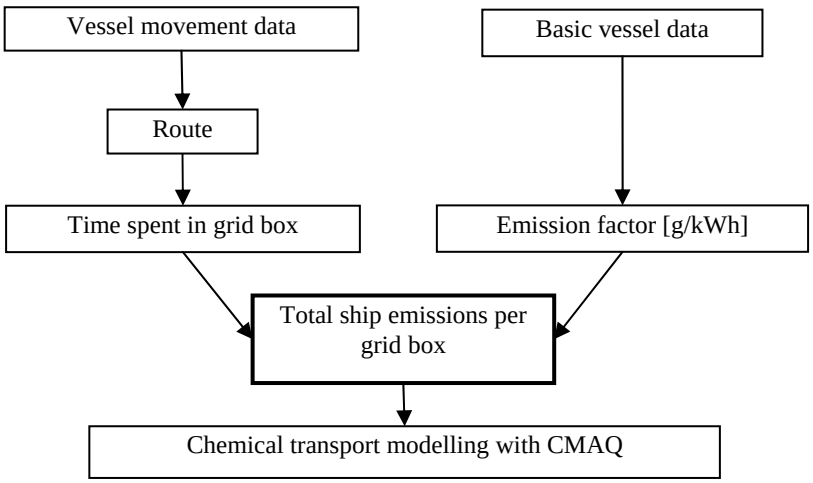

Figure 1: Outline of the procedure to generate the ship emission dataset

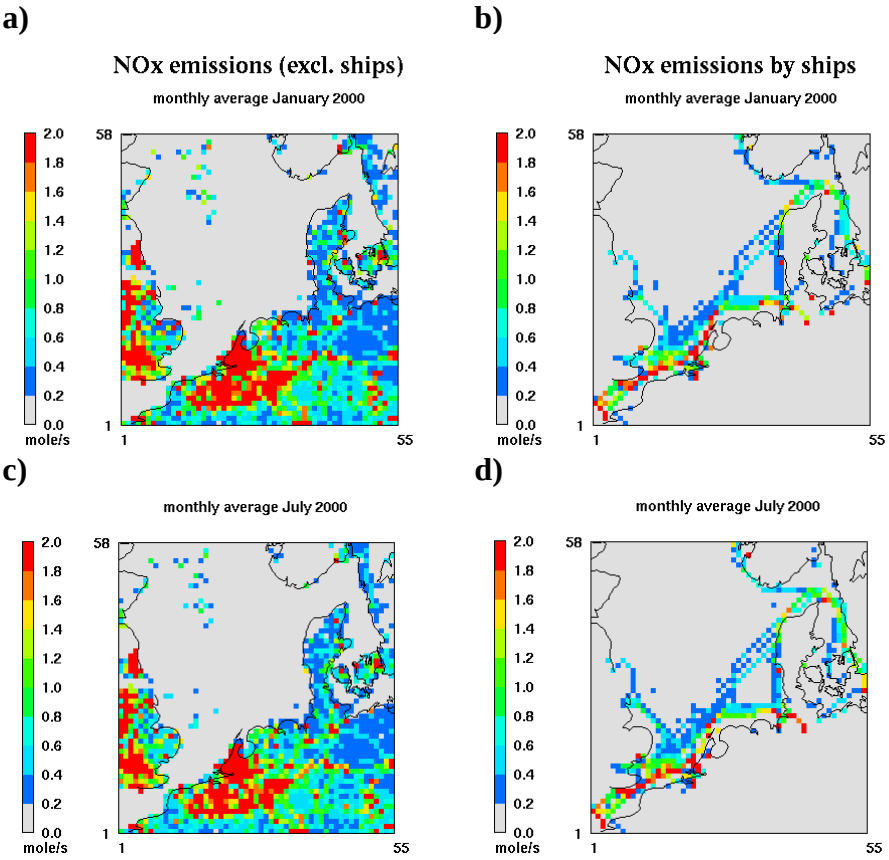

Figure 2: Average land based nitrogen dioxide emissions for January and July 2000 (left side) and the respective ship emissions (right side).

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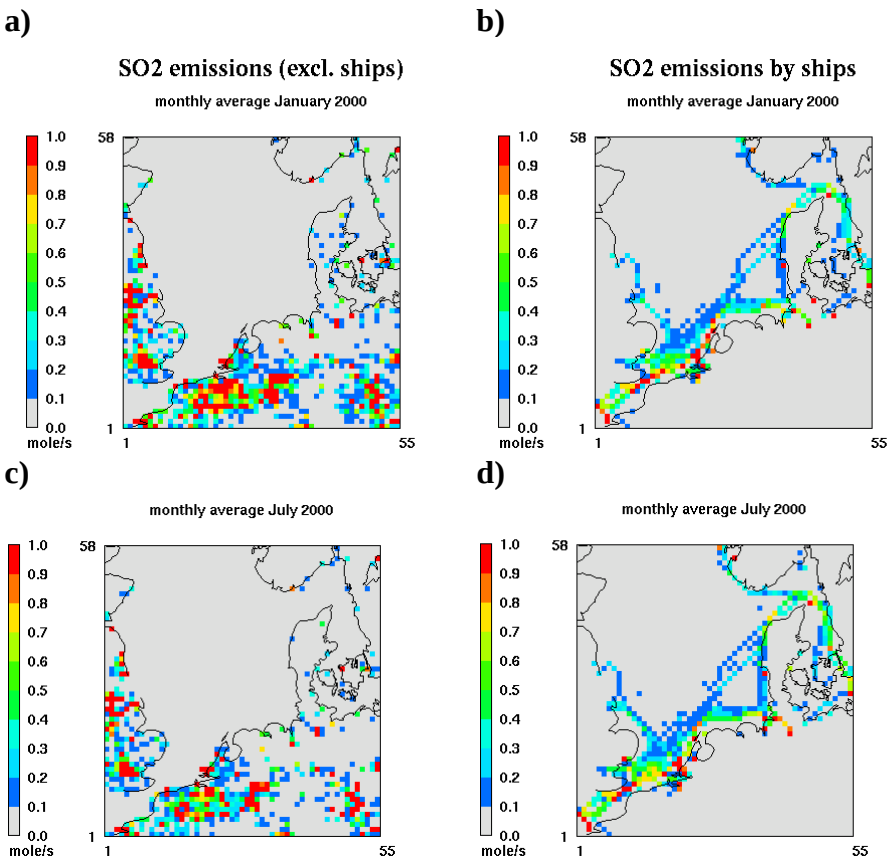

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532 Figure 3: Average land based sulphur dioxide emissions for January and July 2000 (

533 a) and c)) and the respective ship emissions (b) and d)).

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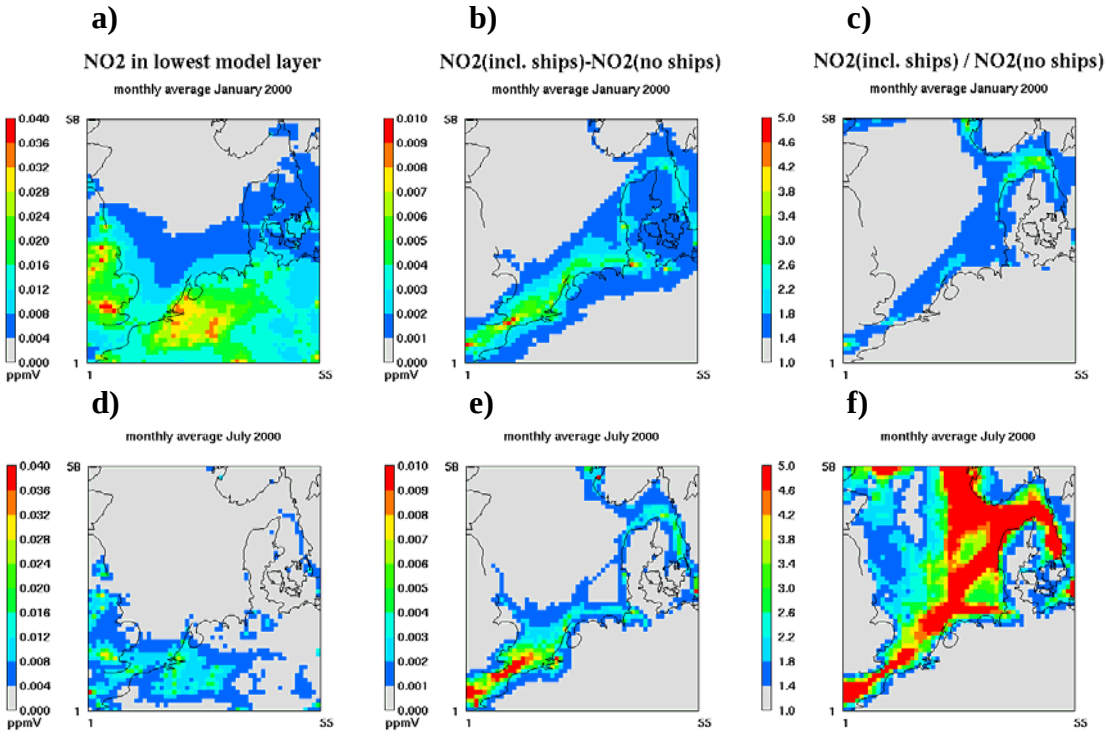


Figure 4: Average nitrogen dioxide concentration for January and July 2000 (a) and d)), the absolute increase caused by ship emissions (b) and e)) and the relative increase caused by ship emissions (c) and f)).

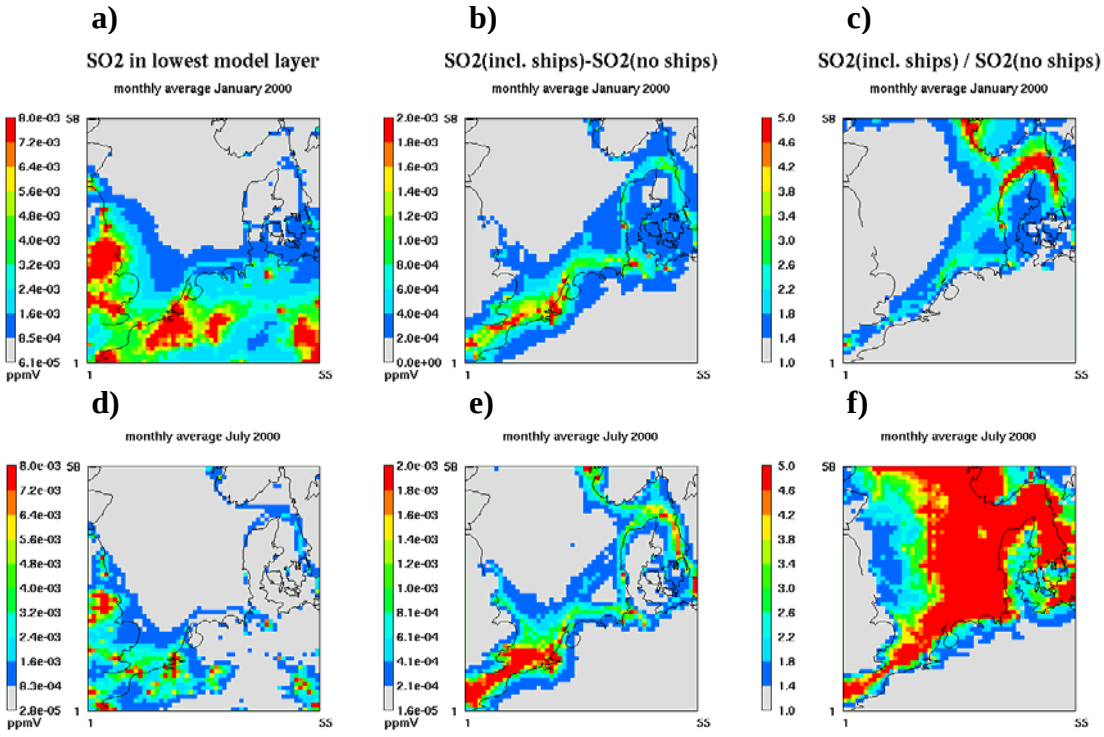
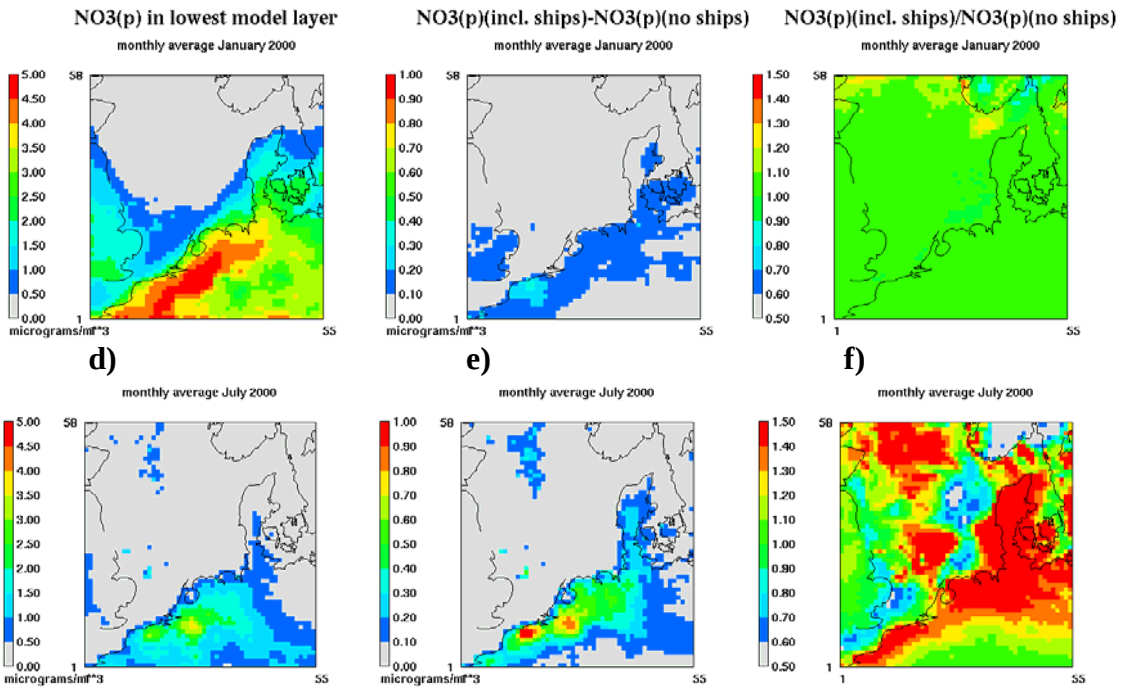


Figure 5: Average sulphur dioxide concentration for January and July 2000 (a) and d)), the absolute increase caused by ship emissions (b) and e)) and the relative increase caused by ship emissions (c) and f)).

549 **a)** **b)** **c)**



550
551 **d)** **e)** **f)**

552
553 Figure 6: Average nitrate aerosol concentration for January and July 2000 (a) and d)

554), the absolute increase caused by ship emissions (b) and e)) and the relative

555 increase caused by ship emissions (c) and f)).

556

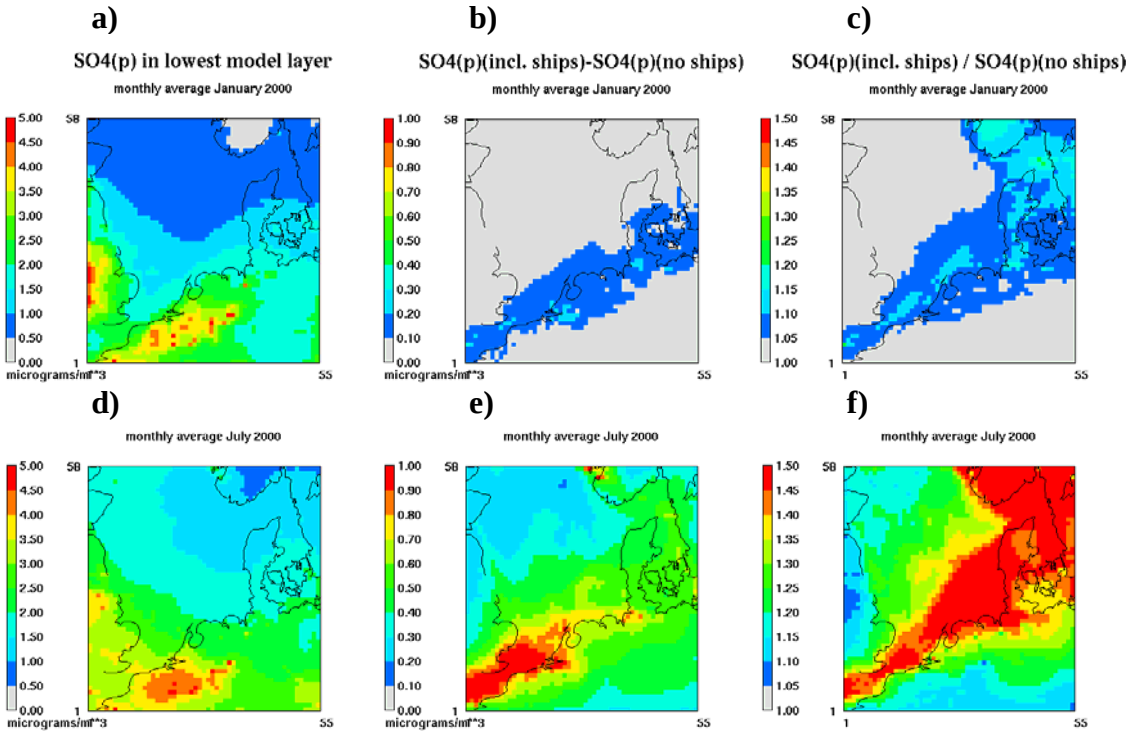
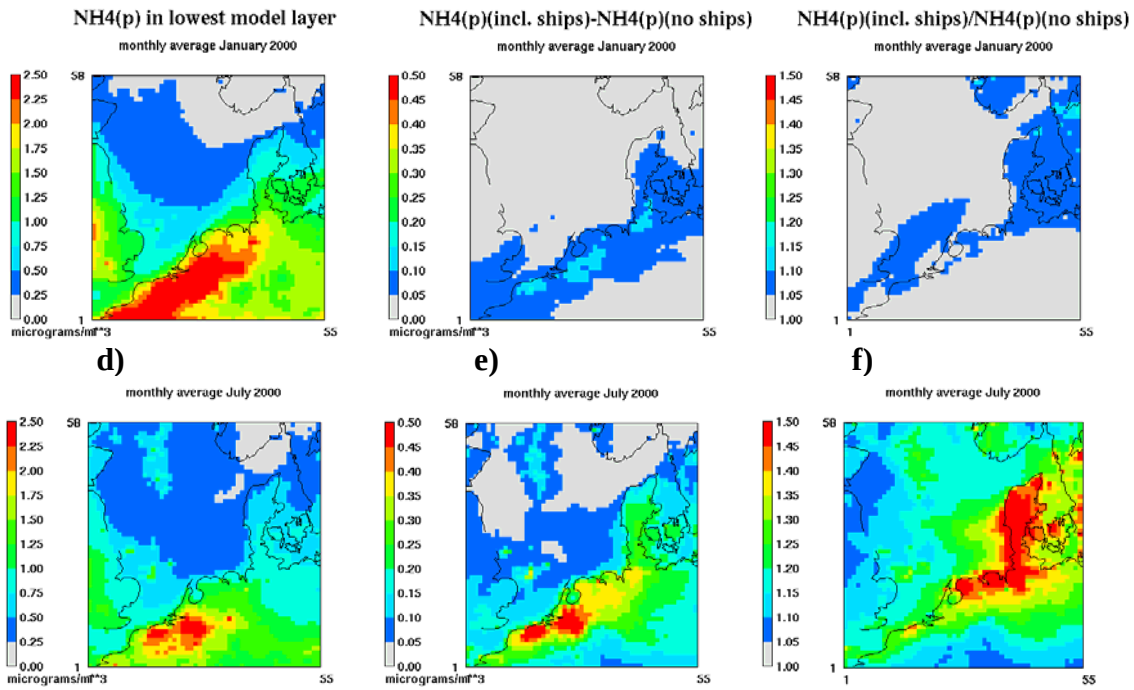


Figure 7: Average sulphate aerosol concentration for January and July 2000 (left side), the absolute increase caused by ship emissions (middle) and the relative increase caused by ship emissions (right side).

564 **a)** **b)** **c)**



565

566 **d)** **e)** **f)**

567

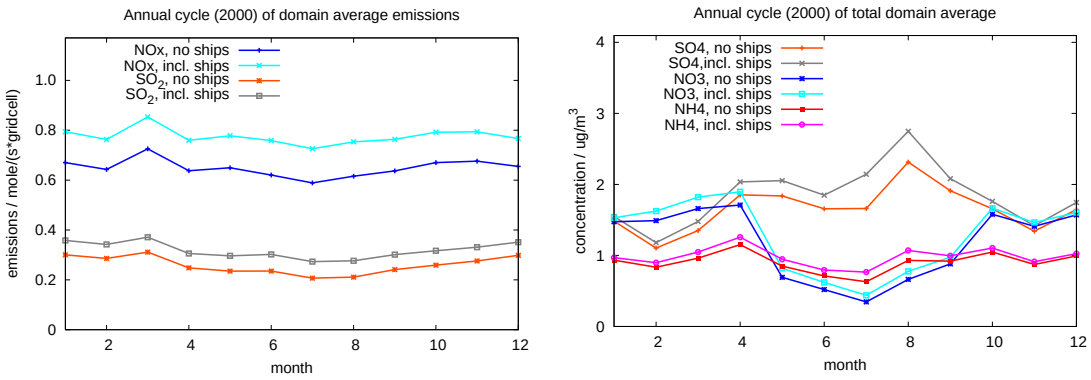
568 Figure 8: Average ammonium aerosol concentration for January and July 2000 (a)

569 and d)), the absolute increase caused by ship emissions (b) and e)) and the relative

570 increase caused by ship emissions (c) and f)).

571

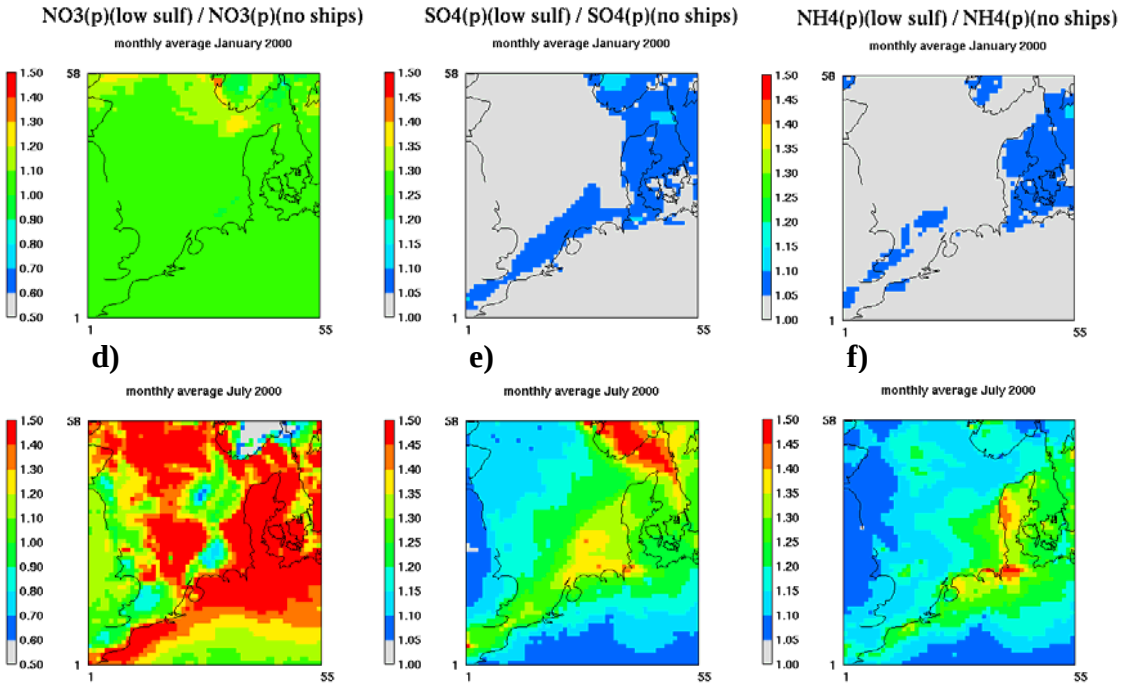
572 **a)** **b)**



573
574 Figure 9: Annual cycle of NO_x and SO_2 emissions (a)) and aerosol concentrations for

575 sulphate, nitrate and ammonium (b)), both averaged over the complete model domain.

576 **a)** **b)** **c)**



577
578 **d)** **e)** **f)**

579

580 Figure 10: Relative increase of nitrate (a) and d)), sulphate (b) and e)) and

581 ammonium (c) and f)) concentrations caused by ship emissions in January and July

582 2000 considering 45 % reduced sulphur content in ship fuels.

583