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# Contribution of ship emissions to the concentration and deposition of air pollutants in Europe

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## Abstract

Emissions from the marine transport sector are one of the least regulated anthropogenic emission sources and contribute significantly to air pollution. Although strict limits were introduced recently for the maximum sulfur content in marine fuels in the SECAs (sulfur emission control areas) and in the EU ports, sulfur emissions outside the SECAs and emissions of other components in all European maritime areas have continued to increase in the last two decades. We have used the air quality model CAMx with and without ship emissions for the year 2006 to determine the effects of international shipping on the annual as well as seasonal concentrations of ozone, primary and secondary components of $PM_{2.5}$ and the dry and wet deposition of nitrogen and sulfur compounds in Europe. Our results suggest that emissions from international shipping affect the air quality in northern and southern Europe differently and their contributions to the air concentrations vary seasonally. The largest changes in pollutant concentrations due to ship emissions were predicted for summer. Increased concentrations of the primary particle mass were found only along the shipping routes whereas concentrations of the secondary pollutants were affected over a larger area. Concentrations of particulate sulfate increased due to ship emissions in the Mediterranean (up to 60 %), in the English Channel and the North Sea (30–35 %) while increases in particulate nitrate levels were found especially in the north, around the Benelux area (20 %) where there were high $NH_3$ land-based emissions. Our model results showed that not only the atmospheric concentrations of pollutants are affected by ship emissions, but also depositions of nitrogen and sulfur compounds increase significantly along the shipping routes. $NO_x$ emissions from the ships especially in the English Channel and the North Sea, cause a decrease in the dry deposition of reduced nitrogen at source regions by moving it from the gas-phase to the particle phase which then contributes to an increase in the wet deposition at coastal areas with higher precipitation. In the western Mediterranean region on the other hand, model results show an increase in the deposition of oxidized nitrogen (mostly $HNO_3$) due to the ship traffic. Dry deposition of





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$SO_2$ seems to be significant along the shipping routes whereas sulfate wet deposition occurs mainly along the Scandinavian and Adriatic coasts. The results presented in this paper suggest that evolution of $NO_x$ emissions from ships and land-based $NH_3$ emissions will play a significant role in the future European air quality.

## 1 Introduction

The rise in population and mobility is associated with emissions of pollutants from transport sectors such as road, air traffic and international shipping. These emissions affect the air quality and climate. There have been many studies on the effects of air and road traffic emissions and projections of their future levels (Cuvelier et al., 2007; Schurmann et al., 2007; Westerdahl et al., 2008; Koffi et al., 2010; Uherek et al., 2010; Wilkerson et al., 2010; Hodnebrog et al., 2011). Relatively few studies, on the other hand, have dealt with the impacts of ship emissions in detail (Corbett et al., 2007; Eyring et al., 2010; Huszar et al., 2010; Jonson et al., 2015).

The marine transport sector, which is one of the least regulated anthropogenic emission sources, contributes significantly to air pollution, particularly in coastal areas (Marmer and Langmann, 2005; Gonzalez et al., 2011). Emissions from maritime transport in European waters constitute a significant share of worldwide ship emissions of air pollutants and greenhouse gases (EEA, 2013). Shipping is one of the fastest growing sources of greenhouse gas emissions due to transport, and is also a major source of air pollution causing health problems, acid rain and eutrophication (Brandt et al., 2013).

Legislation on air pollutants and greenhouse gases from the maritime sector is a major challenge because of the characteristics of the shipping sector, which include global trade operations based in different countries. The efforts of the European Union (EU) and the International maritime Organization (IMO) to tackle emissions from international shipping are different and to date there is no integrated legislation. Globally the International maritime Organization (IMO) is regulating emissions through the Inter-





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national Convention for the Prevention of Pollution from Ships (MARPOL) and its Annex VI, (http://www.imo.org/OurWork/Environment/PollutionPrevention/Pages/Default.aspx). The latest sulfur limits in so-called Emission Control Areas (ECAs) were set at 0.1 % as of 1 January 2015. Reductions of $NO_x$ emissions from marine diesel engines are also regulated, but these focus only on new ships, where limits are defined as a function of speed and installation year.

In Europe, the maximum sulfur content of the marine fuel used by ships operating in the Sulfur Emission Control Areas (SECAs) – the Baltic Sea, the English Channel and the North Sea – was restricted to 1.0 % in July 2010 and further reduced to 0.1 % in January 2015. The EU sulfur directive has limited the sulfur content to 0.1 % in harbor areas since January 2010. Because there is no NECA ($NO_x$ Emission Control Areas) in Europe yet and the IMO $NO_x$ emissions regulations refer only to new ships, the impact of IMO $NO_x$ regulations is minimal at present and probably will continue to be so in the near future (EEA, 2013).

Johansson et al. (2013) reported that the emission limitations from 2009 to 2011 have had a significant effect on reducing the emissions of $SO_x$ in the northern emission control area in Europe. On the other hand, sulfur emissions in sea areas outside the SECAs and emissions of other species – especially $NO_x$ – in all sea areas around Europe have been increasing over the past decades, while land-based emissions have been gradually coming down. The revised Gothenburg Protocol specifies national emission reduction commitments in Europe to be achieved by 2020 (http://www.unece.org/env/lrtap/multi_h1.html). These commitments however are only for land-based sources and do not cover emissions from international shipping. According to the European Environment Agency, emissions of nitrogen oxides from international maritime transport in European waters are projected to increase and could be equal to land-based sources by 2020 (EEA, 2013). In this modeling study, we investigated the impacts of ship emissions on European air quality in detail by analyzing the seasonal and spatial variations of the contributions from the shipping sector to the





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concentrations of ozone and $PM_{2.5}$ components as well as to the deposition of nitrogen and sulfur compounds.

## 2 Method

The models used in this study are the Comprehensive Air quality Model with extensions, CAMx, Version 5.40 (http://www.camx.com) and the Weather Research & Forecasting Model (WRF-ARW), Version 3.2.1 (http://wrf-model.org/index.php). The model domain covered all of Europe with a horizontal resolution of 0.250° × 0.125°. We used 6 h ECMWF data (http://www.ecmwf.int/) to provide initial and boundary conditions for the WRF model. WRF uses 31 vertical layers up to 100 hPa in WRF, of which 14 were used in CAMx with the lowest layer being 20 m thick. The initial and boundary concentrations were obtained from the MOZART global model and adjusted to the CAMx model domain resolution (Horowitz et al., 2003). Photolysis rates were calculated using the TUV photolysis pre-processor (http://cprm.acd.ucar.edu/Models/TUV/). Ozone column densities were extracted from TOMS data (http://ozoneaq.gsfc.nasa.gov). Dry deposition of gases in CAMx is calculated using a state-of-the-science, LAI (leaf-area index)-based resistance model (Zhang et al., 2003). For surface deposition of particles, CAMx includes diffusion, impaction and/or gravitational settling. CAMx uses separate scavenging models for gases and aerosols to calculate wet deposition. The gas-phase mechanism used in this study was CB05 (Carbon Bond Mechanism 5) (Yarwood et al., 2005). Concentrations of particles with a diameter smaller than 2.5 µm were calculated using the fine/coarse option of CAMx. Calculation of secondary organic aerosols (SOA) was based on the semi-volatile equilibrium scheme called SOAP (Strader et al., 1999) that partitions condensable organic gases to seven types of secondary organic aerosols.

The gridded TNO-MACC data for 2006 were used as the basic anthropogenic emission inventory (Denier van der Gon et al., 2010). The annual emission data for 10 SNAP (Selected Nomenclature for sources of Air Pollution) categories per grid cell





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in geographic latitude–longitude coordinate system were converted to hourly, gridded data using the monthly, weekly and diurnal profiles provided by TNO. The biogenic emissions (isoprene, monoterpenes, sesquiterpenes) were calculated as described in Andreani-Aksoyoglu and Keller (1995) using the temperature and shortwave irradiance from the WRF output, the global USGS land use data and the GlobCover 2006 inventory. All emissions were treated as area emissions in the first model layer. We performed CAMx simulations for 2006 with and without ship emissions. Figure S1 shows the annual emissions from ships. Temporal profiles for ship emissions show a small increase (~ 10 %) in summer with respect to winter (Denier van der Gon et al., 2011). Concentrations as well as dry and wet deposition of pollutants were calculated over the entire year. In general, the performance of both WRF and CAMx models was reasonably good for the modeled period with some underestimation of $PM_{2.5}$ during January-February when unusually high concentrations were reported in Europe due to severe meteorological conditions. Details of the model performance of the base run including ship emissions have already been published (Aksoyoglu et al., 2014).

## 3 Results and discussion

### 3.1 Annual impacts

The annual mean surface ozone was predicted to be about 40 ppb over the sea and coastal areas when emissions from the marine transport sector were excluded (Fig. S2). Ship emissions cause an increase in the mean surface ozone by 4–5 ppb (5–10 %) in the Mediterranean Sea (Fig. 1). On the other hand, ozone levels decrease by about 5–6 ppb (10–20 %) around the English Channel and the North Sea due to enhanced titration caused by $NO_x$ emissions from ships. It was shown in an earlier sensitivity study for the same year that ozone formation in that area was VOC-limited because of high $NO_x$ / VOC ratios whereas a $NO_x$-sensitive regime was predicted for the Mediterranean region (Aksoyoglu et al., 2012).





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The modeled mean annual concentration of $PM_{2.5}$ varied between 5 and 40 µg m$^{-3}$ for the year 2006 without ship emissions in Europe (Fig. S3). The highest concentrations were predicted around the Benelux area, northern Italy and eastern Europe. The concentration of $PM_{2.5}$ increased along the shipping routes as well as the coastal areas when emissions from the ship traffic were included (Fig. 2). These changes were caused not only by primary PM (elemental carbon (EC) and primary organic aerosol (POA)) emissions from ships, but also by an increase in the concentration of precursor species leading to the formation of secondary PM (particulate nitrate ($NO_3$), sulfate ($SO_4$), ammonium ($NH_4$) and secondary organic aerosol (SOA)). The largest contribution was predicted in the western Mediterranean (up to 45 %) as well as along the north European coast (10–15 %). Studies with other models using the 2005 inventory at a relatively coarse resolution of about 50 km showed a similar spatial distribution but predicted a lower contribution (15–25 %) in the Mediterranean (EEA, 2013). The difference is probably due to the use of different emission inventories, in addition to the different domain resolutions. The finer resolution used in this study was able to capture the local effects more clearly.

### 3.2 Seasonal impacts

#### 3.2.1 Ozone

We analyzed the changes in the surface ozone mixing ratios caused by the ship emissions in each season separately (Fig. 3). The effects were stronger in summer and there was a difference in the seasonal variation between north and south. Ship emissions were predicted to cause a decrease in ozone in the north, including the area of the English Channel, the North Sea and the Baltic Sea in all seasons except summer. Ozone decreased in summer due to ship traffic only around the English Channel by −20 % while it increased by about 5 % in the eastern part of the North Sea and the Baltic Sea (Fig. 3b). These results are in the same range as those found by Huszar et al. (2010) for 2004. The area around the English Channel is a high-$NO_x$ region lead-





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ing to a reduction of the surface ozone concentration as a result of the contribution from ship emissions as discussed in Sect. 3.1.

On the other hand, an opposite effect was predicted for the southern part of the model domain. Emissions from shipping led to increased surface ozone in all seasons except in winter. No increase, but instead a small decrease, in winter ozone was predicted along the shipping routes (Fig. 3d). In summer, the contribution of the ship emissions to the mean surface ozone varied between +10 and +20 % in the Mediterranean with a negative change of about −5 % over a very small area at the Strait of Gibraltar (Fig. 3b). Marmer et al. (2009) reported the maximum contribution of ships to surface ozone in summer 2006 as 12 % over the Strait of Gibraltar using a global model with a horizontal resolution of 1° × 1°. The finer horizontal resolution used in our study (0.250° × 0.125°) enabled us to distinguish the change in contribution of ship emissions to ozone from +20 % over the northwest African coast to −5 % at the Strait of Gibraltar.

### 3.2.2 $PM_{2.5}$

The model results suggested that emissions from the international shipping increase $PM_{2.5}$ concentrations in all seasons (Fig. 4). The largest contribution of ship traffic was predicted in summer when concentrations increased not only around the shipping routes, but also over the coastal areas. The change in $PM_{2.5}$ concentrations caused by shipping emissions in summer was about 20–25 % in the north around the English Channel and the North Sea, whereas a much larger contribution was predicted in the western Mediterranean (40–50 %). In winter, the contribution decreased to 5–10 % in the north and 15–20 % in the south.

### 3.2.3 Impacts on aerosol components in summer

In this section, the contribution of ship emissions in summer to the individual components of $PM_{2.5}$ is investigated, because the effects are stronger in that season (see Fig. 4b). In order to understand which components are affected more by ship emis-





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sions, we first analyzed the effects on primary and secondary species. The contribution of ship emissions to the concentrations of primary and secondary $PM_{2.5}$ is shown in Fig. 5. Elevated concentrations of the primary carbonaceous components EC and POA were predicted only along the shipping routes in the Mediterranean and in the north around the English Channel and the North Sea (Fig. 5a) whereas the concentrations of secondary aerosols (SA) containing secondary inorganic (SIA) and secondary organic (SOA) aerosols increased over a larger area (Fig. 5b). These results suggest that the effects on the concentrations of secondary particles (via formation by oxidation of gaseous precursors) are more significant than the effects on primary particles (by direct emissions). As seen in Fig. 5b, the concentrations of secondary aerosols increased not only over the sea areas but also over the continent due to emissions from international shipping.

Detailed analysis of model results revealed that the change in the secondary aerosol concentration due to ship emissions occurs mainly in the inorganic fraction (Fig. 6a–c). The concentrations of particulate nitrate and ammonium increased by about 10–20 % around the Benelux area and northern Italy where there are high land-based ammonia emissions (Fig. 6a and b). These results indicate that $NO_x$ emissions from the ships and ammonia emissions from the land lead to the formation of ammonium nitrate. On the other hand, particulate sulfate increased along the shipping routes and coastal areas with the largest effects (50–60 %) in the western Mediterranean and the North African coast (Fig. 6c). The contribution to the SOA concentration was relatively small (< 10 %) and was mainly found in the north (Fig. 6d). We note that the results for SOA might look different if a VBS (Volatility Basis Set) scheme were used to calculate the organic aerosol (OA) concentrations (Donahue et al., 2006), but this could not be done because the volatility distribution of ship emissions is not well known yet (Pirjola et al., 2014).





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### 3.3 Contribution to deposition

#### 3.3.1 Nitrogen deposition

The atmospheric deposition of pollutants raises serious concerns for ecosystems. In general, the main nitrogen sources are emissions of nitrogen oxides from combustion processes and ammonia from agricultural activities. The deposition of atmospheric nitrogen species constitutes a major nutrient input to the biosphere, which enhances forest growth. Despite this, increased nitrogen input into terrestrial ecosystems represents a potential threat to forests. Enhanced nitrogen deposition can cause soil acidification, eutrophication and nutrient imbalances, causing a reduction in biodiversity. The deposition of atmospheric nitrogen compounds occurs via dry and wet processes. $NO_2$, $NH_3$, nitric acid ($HNO_3$), and nitrous acid (HONO) are the most important contributors to nitrogen dry deposition. Nitrogen wet deposition results from the scavenging of atmospheric N constituents.

The predicted annual deposition of total nitrogen in Europe based only on the land emissions varied between 5 and 45 kg N $ha^{-1}$ $yr^{-1}$ in 2006 (Fig. 7, left) and it was mainly dominated by dry deposition (Fig. S4). The largest dry deposition was generally over the regions with high ambient $NH_3$ concentrations (the Benelux area and northern Italy) as also reported previously (Flechard et al., 2011). In the rest of the area, dry deposition of oxidized nitrogen dominated.

As seen in the right panel of Fig. 7, ship emissions caused an increase in N deposition along the shipping routes except for a few high-$NH_3$ locations where a small decrease in deposition was predicted. Analysis of the changes in the dry and wet deposition showed that the main contribution of ship emissions was to dry N deposition while wet deposition increased slightly (10 %) in the North Sea (Figs. 8 and S5).

Further investigation of the changes in the dry deposition showed that ship emissions caused an increase in the dry deposition of $HNO_3$ in the Mediterranean whereas there was a small decrease (−2 %) in the $NH_3$ deposition in ammonia-rich areas (Fig. 9). Dry deposition of ammonia occurred close to the source areas. Our results suggest





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that $NO_x$ emissions from ships were responsible for transformation of some gaseous ammonia to particulate ammonium (see Fig. S6), which has a lower dry deposition velocity than gaseous $NH_3$ but contributes to an increased wet deposition especially over the North Sea (Fig. 8, right panel). The largest contribution of the ship traffic emissions to deposition of oxidized nitrogen (in the form of $HNO_3$) was in the Mediterranean Sea (see Fig. 9, right panel).

### 3.3.2 Sulfur deposition

After emission, sulfur dioxide is further oxidized in the atmosphere, with sulfuric acid and sulfate as final products. Sulfate is mostly removed by wet deposition, with various effects on ecosystems including acidification of marine ecosystems and soil, vegetation damage, as well as corrosion. Excluding the ship emissions, the largest total sulfur deposition was predicted to occur in the eastern part of Europe (with high land-based $SO_2$ emissions) (Fig. 10, left) and was dominated by dry deposition (Fig. S7, left panel). Wet deposition was predicted to be relatively higher in areas with high precipitation (Fig. S7, right panel). Generally, the importance of dry deposition of sulfur decreased and the importance of wet deposition increased with distance from the source, along with the decrease of the $SO_2$ / sulfate ratio.

Our simulations showed that ship emissions contributed substantially to the sulfur deposition along the shipping routes and the coastal areas (Fig. 10, right panel, see Fig. S8 for relative contribution). The western Mediterranean and the North African coast were especially affected by the sulfur deposition from ship traffic. As shown in Fig. 11, the contribution to the dry $SO_2$ deposition dominated along the shipping routes while the effect on wet $SO_4$ deposition was smaller and was mostly in areas with higher precipitation. Comparison of the right panel of Fig. 10 with the left panel of Fig. 11 shows clearly that the contribution of ship emissions to sulfur deposition is mainly as $SO_2$ dry deposition.





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## 4 Conclusions

Although regulations for emissions from the maritime traffic sector – especially for sulfur – have been tightened over the last few years, the impacts are limited at present in Europe since there is no NECA ($NO_x$ Emission Control Area) yet and the IMO emissions limits refer only to new ships. The European Environment Agency estimated that emissions of nitrogen oxides from international maritime transport in European waters could be equal to land-based sources by 2020. The model results presented in this study give an overview of the effects of ship emissions on the concentrations and depositions of air pollutants in Europe, based on the 2006 emission inventory.

Our results suggest that emissions from marine engines cause a decrease of 10–20 % in annual surface ozone in the area of the English Channel and the North Sea, but they lead to an increase (5–10 %) in the Mediterranean Sea. There was a difference in the seasonal variation between north and south. Ship emissions were predicted to cause a decrease in ozone in the north covering the area of the English Channel, the North Sea and the Baltic Sea in all seasons except summer. Ozone decreased in summer due to ship traffic only around the English Channel while it increased by about 5 % in the North and the Baltic Seas. On the other hand, an opposite effect was predicted for the southern part of the model domain. Emissions from shipping led to an increase in the surface ozone in all seasons except in winter. In contrast, a small decrease in winter ozone was predicted along the shipping routes especially in the western Mediterranean. Based on these results, we conclude that ship emissions cause an increase in ozone in seasons with active photochemistry (i.e. summer in the north and spring to fall in the south).

The $PM_{2.5}$ concentrations increased by up to 45 % in the Mediterranean Sea, and 10–15 % in the North Sea, Baltic Sea and along the coastal areas due to ship traffic. The impacts predicted for the Mediterranean region are larger than those reported in other studies. The finer resolution used in this work captured the local effects more accurately. Significant effects of ship emissions on the air quality were predicted not





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only along the shipping routes, but also over a large part of the European continent. Although increased concentrations of primary organic aerosols and elemental carbon were predicted only along the shipping routes, secondary pollutants were affected over a larger area. The effects of ship emissions were larger in summer predominantly on secondary inorganic aerosols whereas secondary organic aerosol concentrations increased by less than 10 %. Ship emissions increased the particulate sulfate concentrations in the Mediterranean as well as in the North Sea. On the other hand, particulate nitrate concentrations increased due to the $NO_x$ emissions from shipping, especially around the Benelux area where there are high land-based $NH_3$ emissions.

Consumption of gaseous $NH_3$ for particulate nitrate formation resulted in a small decrease in the dry deposition of reduced nitrogen in its source regions and an increase in wet deposition along the shorelines with high precipitation rates. Deposition of nitrogen was predicted to increase in the Mediterranean mainly due to an increase in the deposition of oxidized nitrogen compounds (mainly $HNO_3$). On the other hand, the increase in dry deposition of $SO_2$ along the shipping routes was larger than the increase in wet deposition of $SO_4$ along the Scandinavian and the Adriatic coast.

The model results achieved in this study suggest that emissions from ship traffic have significant impacts on air quality, not only along the shipping routes but also over a large part of the European continent. While $SO_2$ emissions in European waters will continue to decrease due to regulation of the sulfur content in marine fuels, $NO_x$ emissions are expected to increase further in the future and could be equal to or even larger than the land-based emissions from 2020 onwards. Impacts of regulations for $NO_x$ emissions from marine diesel engines are expected to be limited in the near future.

In an earlier study, we predicted that there would be a significant reduction in $PM_{2.5}$ ($\sim$ 30 %) and in oxidized nitrogen deposition ($\sim$ 40 %) in Europe by 2020 by comparison with 2005, assuming a baseline scenario where land-based emissions were reduced according to the Gothenburg Protocol Scenarios (Aksoyoglu et al., 2014). Increasing emissions from the marine transport, however, might partly outweigh the benefit from reductions of land-based emissions.





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As a final remark, we have to consider the following issues for future European air quality: with significant future reductions of $NO_x$ emissions from ship traffic, changing chemical regimes around the northern coast would affect the impacts on ozone as well as the formation of secondary inorganic aerosols. Decreasing $NO_x$ / VOC ratios would affect ozone formation whereas decreasing $NO_x$ / $NH_3$ ratios might change the formation of secondary inorganic aerosols as well as nitrogen deposition since ammonia land emissions are not expected to decrease significantly in the near future compared to sulfur and nitrogen emissions in Europe.

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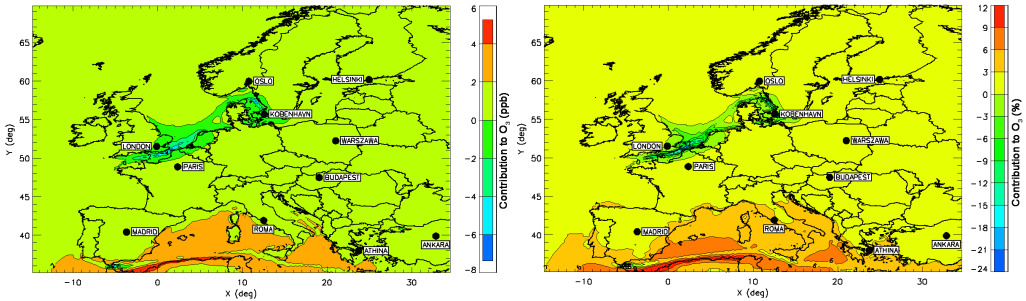


**Figure 1.** Contribution of ship emissions (left in ppb, right in %) to mean surface $O_3$ in 2006.





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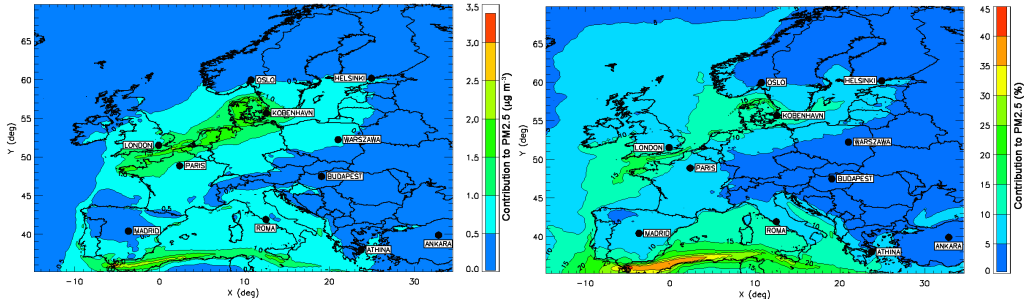


**Figure 2.** Contribution of ship emissions (left in $\mu g\,m^{-3}$, right in %) to the mean $PM_{2.5}$ concentration in 2006.





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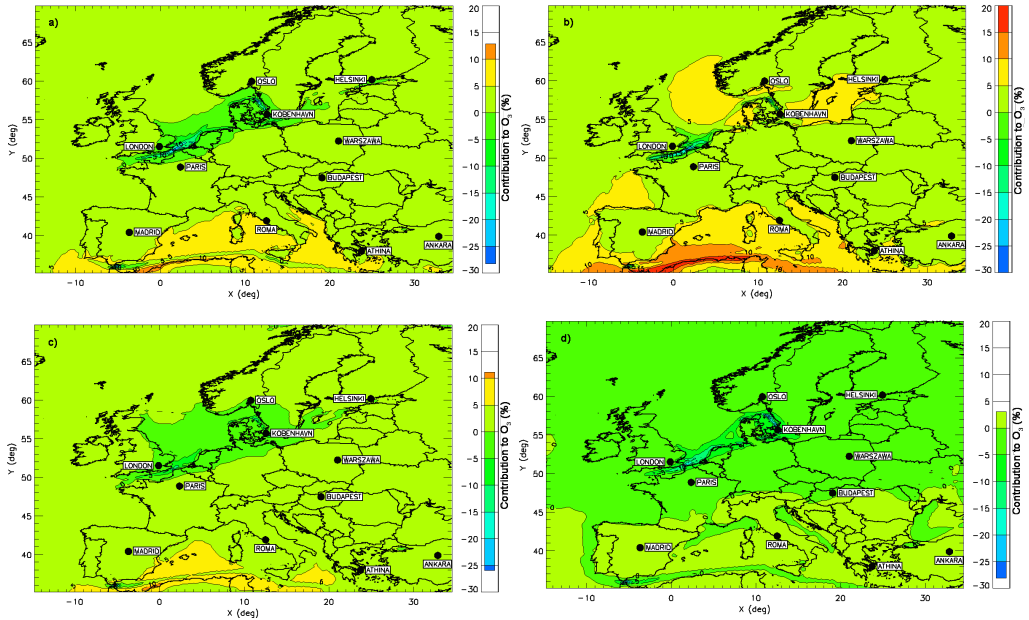


**Figure 3.** Contribution of ship emissions to mean surface $O_3$ (%) in **(a)** spring, **(b)** summer, **(c)** fall, and **(d)** winter.





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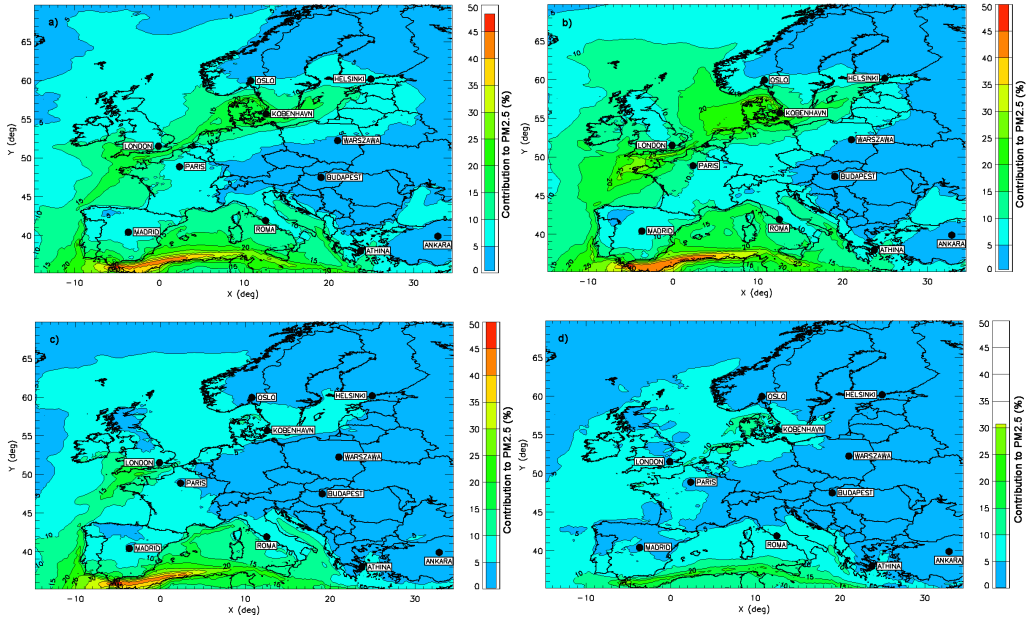


**Figure 4.** Contribution of ship emissions to $PM_{2.5}$ (%) in **(a)** spring, **(b)** summer, **(c)** fall, and **(d)** winter.





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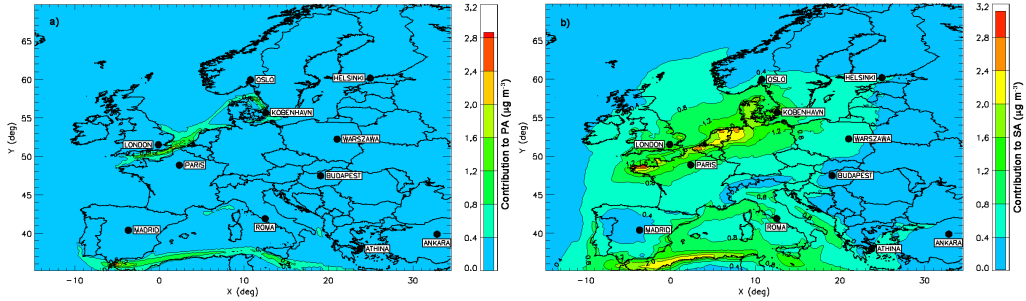


**Figure 5.** Contribution of ship emissions ($\mu g\,m^{-3}$) to **(a)** the primary aerosol (PA), **(b)** the secondary aerosol (SA) concentration in summer 2006.





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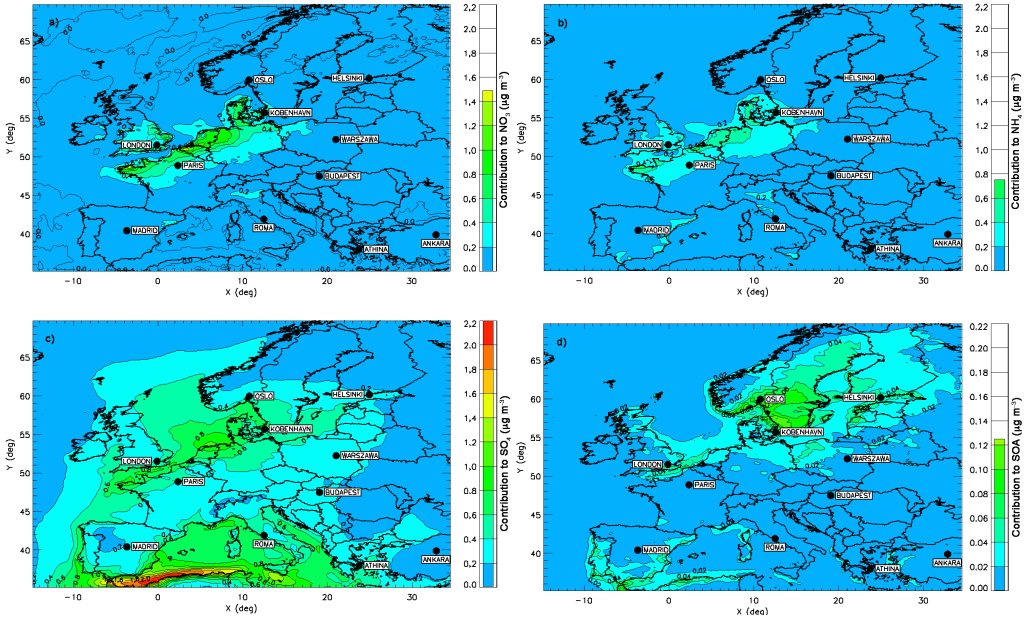


**Figure 6.** Contribution of ship emissions ($\mu g\,m^{-3}$) to the secondary aerosol concentration; **(a)** $NO_3$ **(b)** $NH_4$ **(c)** $SO_4$ **(d)** SOA in summer 2006. Note that the scale in **(d)** is ten times smaller than the others.





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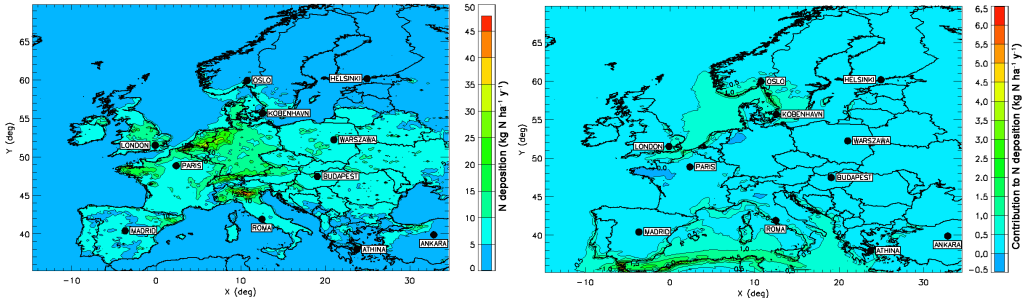


**Figure 7.** Annual nitrogen deposition only due to land-based emissions (left) and contribution of ship emissions to N deposition (right).





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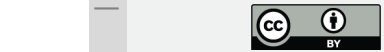

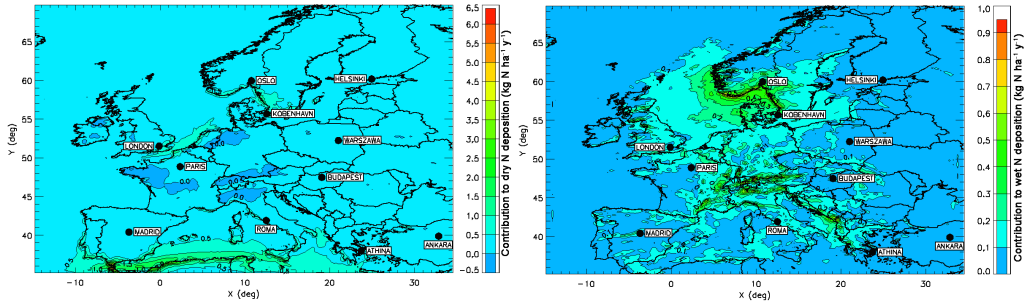


**Figure 8.** Contribution of ship emissions to the annual dry N deposition (left) and wet N deposition (right).





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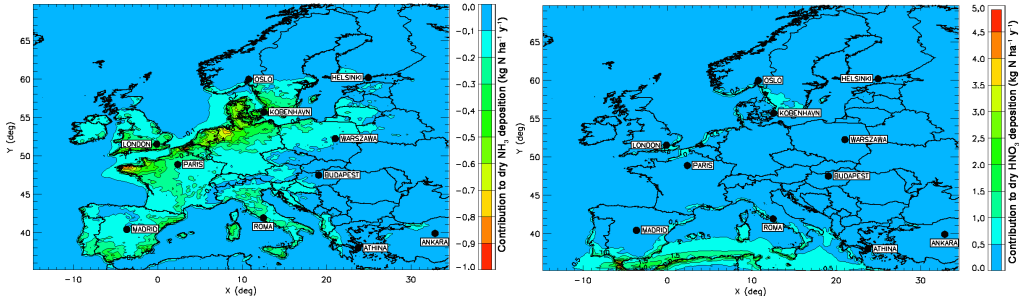


**Figure 9.** Contribution of ship emissions to the annual dry $NH_3$ deposition (left) and dry $HNO_3$ deposition (right).





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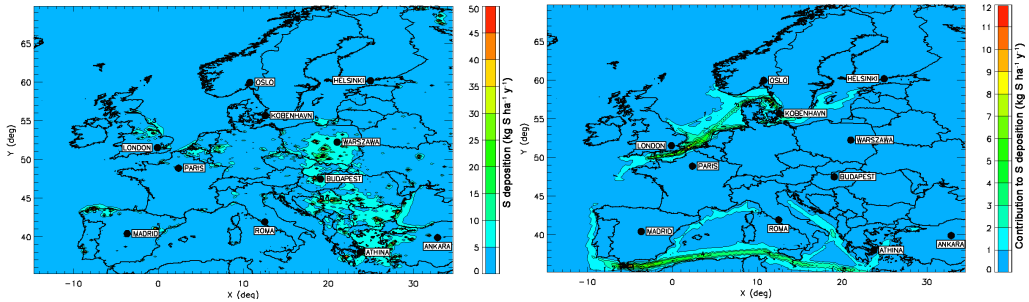


**Figure 10.** Sulfur deposition only due to land-based emissions (left) and due to ship emissions (right).





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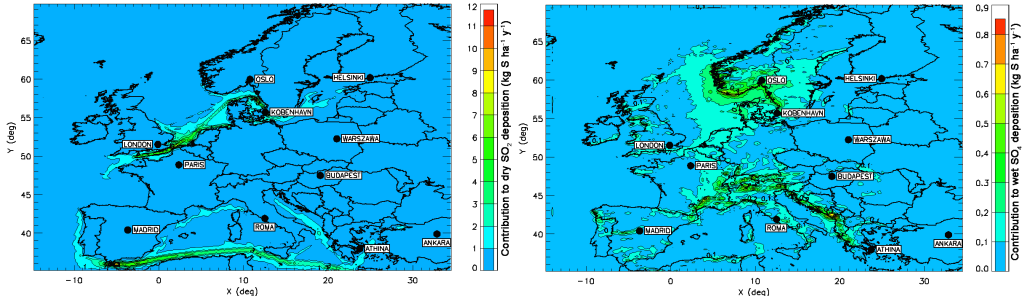


**Figure 11.** Contribution of ship emissions to dry $SO_2$ deposition (left) and wet $SO_4$ deposition (right).





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