

1 **Reply to comments of Anonymous Referee # 1:**

2 Original comments are in **black**, replies in **blue** and proposed new text is in *italic*.

3 This manuscript presents valuable results on the influence of shipping on the  
4 concentrations and deposition in Europe, for selected pollutants. The results are  
5 worth publishing; however, the following comments for improving the manuscript first  
6 need to be taken into account.

7 **Thank you for your comments and suggestions to improve our manuscript. Our**  
8 **answers to specific questions follow below:**

9 Major comments

10 The use of shipping AIS signals in emission modelling has facilitated major  
11 improvements regarding the accuracy on how the shipping emissions can be treated.  
12 This is a key issue in terms of the aims and contents of this study. The authors  
13 should therefore properly discuss these new developments in the introductory  
14 section.

15 **We fully agree about the importance of new technologies such as AIS (Automatic**  
16 **Identification System). The further development of shipping monitoring tools, such as**  
17 **AIS and availability of data generated with these monitoring tools, will allow a better**  
18 **estimation of shipping emissions. We added the following text in the introduction:**

19 *The highest level of detail on ship movements can be obtained with the AIS*  
20 *(Automatic Identification System) data. The AIS was developed and made*  
21 *compulsory by the International Maritime Organization for all ships over 300 gross*  
22 *tonnage to minimize the probability of groundings and collisions of ships. These*  
23 *signals allow very accurate positioning of vessels and their emissions. When*  
24 *combined with knowledge on each ship's engine and possible abatement techniques,*  
25 *a realistic estimation of fuel consumption and emissions can be made. Jalkanen et al.*  
26 *(2009) presented an automated system that is based on AIS signals, to evaluate*  
27 *exhaust emissions from marine traffic in the Baltic Sea area. A pilot project using the*  
28 *AIS data to estimate shipping emissions in the port of Rotterdam allowed for*  
29 *calculation of emissions on a much finer geographical grid than could be done*  
30 *previously (Denier and Hulskotte, 2010). In the near future, AIS data is expected to*  
31 *be used to improve accuracy of emission estimates in a larger area in Europe.*

1 The authors should also present in the manuscript a proper evaluation of the  
2 accuracy of their numerical results. How accurate is the emission inventory for  
3 various source categories? How accurate are the predictions of their chemical  
4 transport modeling (CTM)? What are the most significant uncertainties of the  
5 emission inventory and the CTM? For instance, what is known of the accuracy of  
6 modelling biogenic emissions, secondary organics, dry and wet deposition?

7 The model performance for simulations reported in this paper was thoroughly  
8 evaluated and the results were published in Aksoyoglu et al. (2014). Accuracy of the  
9 state-of-the-art air quality models such as CAMx, depends largely on the quality of  
10 the input data such as meteorological fields and emissions. It is well known that  
11 reproducing the meteorological parameters like wind fields under difficult conditions –  
12 especially in wintertime- is challenging. As shown in Aksoyoglu et al. (2014), results  
13 of meteorological model WRF for this application were quite satisfactory.

14 Emissions are another very important input for CTMs. Anthropogenic emissions are  
15 based on the reported data by countries and uncertainties are mostly related to lack  
16 of some information such as wood burning emissions. In this work, we used TNO-  
17 MACC European emission inventory which has been applied in several European  
18 modeling projects (Denier van der Gon et al., 2010, Pouliot et al., 2012). Uncertainty  
19 in emissions varies depending on pollutant and source (Kuonen et al., 2014). In  
20 general, some emission sources are difficult to estimate regionally, such as fugitive  
21 dust and agricultural activities. For example, ammonia emissions are dominated by  
22 agricultural operations and their daily and diurnal variations are related to actual  
23 climate conditions in a particular year.

24 Biogenic emission models require a detailed vegetation inventory, emission factors  
25 (based on a very few data) for each specific species as well as temperature and  
26 radiation data (Guenther et al. 2012, Oderbolz et al., 2013). In spite of extensive  
27 efforts in this field, biogenic emission models still have high uncertainty mostly due to  
28 lack of sufficient measurements of these species.

29 Modeling of secondary organic aerosols has been the focus of substantial research  
30 worldwide, since Robinson et al. (2007) reported the semi-volatile character of  
31 primary emissions. Many CTMs have already implemented the Volatility Basis Set  
32 (VBS) developed by Donahue et al. (2011) to improve SOA modeling.

1 Evaluation of modeled deposition is more challenging since measurement techniques  
2 are available only for wet deposition. Dry deposition can only be estimated using gas  
3 phase concentrations and dry deposition velocities.

4 We added the following text in page 30964, line11:

5 *Model performance and uncertainties:*

6 *The model performance for simulations reported in this paper was thoroughly*  
7 *evaluated and the results were published in Aksoyoglu et al. (2014). It is however,*  
8 *necessary to give some information about the model performance here. Accuracy of*  
9 *the state-of-the-art air quality models such as CAMx, depends largely on the quality*  
10 *of the input data such as meteorological fields and emissions. It is well known that*  
11 *reproducing the meteorological parameters like wind fields under difficult conditions –*  
12 *especially in wintertime- is challenging. Uncertainty in emissions varies depending on*  
13 *pollutant and source. In general, some emission sources are difficult to estimate*  
14 *regionally, such as agricultural activities. For example, ammonia emissions and their*  
15 *daily and diurnal variations are related to actual climate conditions in a particular*  
16 *year. According to Kuonen et al. (2014), uncertainty estimates for emissions vary*  
17 *between 10-300% depending on pollutant and source.*

18 *Biogenic emission models require a detailed vegetation inventory, emission factors*  
19 *(based on a very few data) for each specific species as well as temperature and*  
20 *radiation data (Guenther et. al. 2012, Oderbolz et al., 2013). In spite of extensive*  
21 *efforts in this field, biogenic emission models still have high uncertainty mostly due to*  
22 *lack of sufficient measurements of these species. Evaluation of deposition is another*  
23 *challenge since measurement techniques are available only for wet deposition. Dry*  
24 *deposition can only be estimated using gas phase concentrations and dry deposition*  
25 *velocities.*

26 *By keeping these uncertainties in mind, the general performance of both WRF and*  
27 *CAMx models was reasonably good for the modeled period with some*  
28 *underestimation of PM<sub>2.5</sub> during January-February when unusually high*  
29 *concentrations were reported in Europe due to severe meteorological conditions. The*  
30 *agreement between measurements and meteorological model results was good, with*  
31 *high correlation coefficients (0.76–0.98) and low mean bias error, MBE (-1.13 for air*  
32 *temperature, 0.57 for wind speed). These values fulfil the desired accuracy*

1 suggested by Cox et al. (1998). The model evaluation of the CAMx model suggested  
2 a relatively good model performance with a mean bias of 4 ppb and  $-1.9 \mu\text{g m}^{-3}$  for  
3 ozone and  $\text{PM}_{2.5}$  concentrations, respectively. Details of the model performance of  
4 the base run including ship emissions have been published in Aksoyoglu et al.  
5 (2014).

6 The considerations on modelling uncertainties should be taken into account in the  
7 interpretation and discussion of the numerical results. The main factors causing  
8 uncertainties should also be discussed in the conclusions section.

9 We added the following in page 30971, line 4:

10 *The effects of ship emissions were larger in summer predominantly on secondary*  
11 *inorganic aerosols whereas secondary organic aerosol concentrations increased by*  
12 *less than 10 %. One should keep in mind however, that the results for SOA might*  
13 *look different if a VBS (Volatility Basis Set) scheme is used to calculate the organic*  
14 *aerosol (OA) concentrations, but this could not be done in this study due to the lack*  
15 *of volatility distribution of ship emissions.*

16 We added the following in page 30972, line 1:

17 *As a final remark, we have to consider the following issues for future European air*  
18 *quality: in general, there is a clear need to improve the emission inventories to*  
19 *reduce the uncertainties; since ammonia is a very important precursor for the*  
20 *secondary inorganic aerosol formation, more accurate estimates of its emissions are*  
21 *needed for future simulations; with significant future reductions of NO<sub>x</sub> emissions*  
22 *from ship traffic, changing chemical regimes around the northern coast would affect*  
23 *the impacts on ozone as well as the formation of secondary inorganic aerosols.*

24 The authors describe their methods, regarding the MACC and biogenic emissions.  
25 However, they should also clearly state, which emission categories were NOT  
26 include- that is good scientific practice. As MACC includes only anthropogenic  
27 emissions, they probably neglected at least wild fire, sea salt and dust emissions. If  
28 all of these were neglected, they should at least provide some estimate (using proper  
29 references) on how large a fraction of emissions for each relevant pollutant was not  
30 taken into account. The neglected source categories have a direct influence on the

1 contribution percentages of shipping, compared with total concentrations and  
2 depositions.

3 The emission inventory used in this study did not have wild fire, sea salt and dust  
4 emissions. There are some estimates of fires using the fire radiative power (FRP)  
5 from MODIS equipped satellites (Sofiev et al., 2013). Occurrence and intensity of  
6 such emissions as well as vertical distributions however, vary significantly spatially  
7 and temporally making their parameterization difficult. In order to avoid further  
8 uncertainties and likely errors, we decided not to include fire emissions in our  
9 simulations, until reliable data parameterization is available.

10 Sea salt modeling has large uncertainties mainly in generation of sea spray which  
11 occurs as the waves break on the surface of the ocean and whitecaps form (Tsyro et  
12 al., 2011). However, sea salt is mainly found on coarse particles and sea salt  
13 modeling would improve mainly formation of coarse nitrate (Sellegri et al., 2001).  
14 Similarly, mineral dust is more relevant for coarse particles (Athanasopoulou et al.,  
15 2010). Since our focus in this work was only on the fine fraction of particles ( $PM_{2.5}$ ),  
16 we believe that lack of such emissions did not affect our results significantly.

17 We added the following comments in the Methods section:

18 *page 30963, line 26: The annual emission data for 10 SNAP (Selected Nomenclature*  
19 *for sources of Air Pollution) categories per grid cell in geographic latitude–longitude*  
20 *coordinate system were converted to hourly, gridded data using the monthly, weekly*  
21 *and diurnal profiles provided by TNO. Wild fire, sea salt and mineral dust emissions*  
22 *were not included in the inventory. There are some estimates of fires using the fire*  
23 *radiative power (FRP) from satellites (Sofiev et al., 2013). Occurrence and intensity*  
24 *of such emissions as well as vertical distributions however, vary significantly spatially*  
25 *and temporally making their parameterization difficult. Sea salt is mainly found on*  
26 *coarse particles and sea salt modeling would improve mainly formation of coarse*  
27 *nitrate (Sellegri et al., 2001). Similarly, mineral dust is more relevant for coarse*  
28 *particles (Athanasopoulou et al., 2010). Since our focus in this work was only on the*  
29 *fine fraction of particles ( $PM_{2.5}$ ), we believe that lack of such emissions did not affect*  
30 *our results significantly.*

31 It should also be reported what was the spatial resolution of the emission inventory  
32 (in kilometers), especially regarding the shipping emissions. The authors should also

1 report the resolution of their chemical transport modelling (CTM) not only in terms of  
2 degrees; but for readability, also report what these correspond as kilometers in the  
3 domain used.

4 Both anthropogenic emissions and models in this study use the geographic  
5 coordinate system (latitude, longitude). Since the size of grid cells varies with the  
6 latitude, one cannot give a grid cell resolution in kilometers. One can however, define  
7 a range in km. We added the following statements:

8 *Page 30963, line 6: The model domain covered all of Europe with a horizontal*  
9 *resolution of  $0.250^\circ \times 0.125^\circ$  which corresponds approximately to 19 km x 13 km*  
10 *around the central latitudes of the model domain.*

11 *Page 30963, line 26: The annual emission data for 10 SNAP (Selected Nomenclature*  
12 *for sources of Air Pollution) categories per grid cell in geographic latitude–longitude*  
13 *coordinate system (with a grid resolution of  $0.125^\circ \times 0.0625^\circ$  which corresponds*  
14 *approximately to 9 km x 7 km around the central latitudes of the model domain) were*  
15 *converted to hourly, gridded data using the monthly, weekly and diurnal profiles*  
16 *provided by TNO.*

17 Minor comments

18 Abstract. “Our results suggest that emissions from international shipping affect the air  
19 quality in northern and southern Europe differently and their contributions to the air  
20 concentrations vary seasonally.” The former part of this sentence is vague  
21 (‘differently’, not stated in which respect), and the latter part is trivial. Remove or  
22 clarify the former part, and delete the latter.

23 Deleted

24 “Increased concentrations of the primary particle mass were found only along the  
25 shipping routes whereas concentrations of the secondary pollutants were affected  
26 over a larger area.” Trivial statement, to be removed.

27 Removed

28 Introduction. “The rise in population and mobility is associated with emissions of  
29 pollutants from transport sectors such as road, air traffic and international shipping.

1 These emissions affect the air quality and climate.” Trivial statements, to be  
2 removed.

3 **Removed**

4 International Maritime Organisation: Maritime is written with a capital letter.

5 **corrected**

6 ‘latest Sulphur limits’, better written as latest fuel Sulphur limits

7 **corrected**

8 Line 9. WRF occurs once too many

9 **corrected**

10 p. 30967, lines 7-10. How much more important are the effects of secondary  
11 compared with primary ? Please state quantitatively.

12 **Significance is not due to quantitative contribution but because of extension of**  
13 **influence over the continental area.**

14

15 **Reply to comments of Anonymous Referee # 2:**

16 Original comments are in **black**, replies in **blue** and proposed new text is in *italic*.

17 The present study uses a CTM to calculate the annual, seasonal and spatial impacts  
18 of shipping emissions in the European waters to ozone and fine particle levels and  
19 composition. Background is very clear but the motivation and aim should be detailed  
20 further. What is the expected outcome of this modelling exercise? Similar studies  
21 have been done before and the impacts are more or less known. On the other hand it  
22 is an advantage to use a finer resolution to capture more local impacts and the study  
23 focuses on the impact on organic and inorganic composition as well as dry and wet  
24 deposition.

25 **Thank you for your comments and suggestions. In order to clarify the motivation of**  
26 **this study we added the following text in the introduction:**

27 *Page 30962, line 11: Although more stringent NOx emission limits legislated by the*  
28 *International Maritime Organization (IMO) have forced marine diesel engine*  
29 *manufacturers to consider a variety of different emission reduction technologies,*

1 *there is no NECA (NO<sub>x</sub> Emission Control Areas) in Europe yet. Since the IMO NO<sub>x</sub>*  
2 *emissions regulations refer only to new ships, the impact of these regulations is*  
3 *minimal at present and probably will continue to be so in the near future (EEA, 2013).*

4 *Line 26: It is therefore important to understand the impacts of shipping emissions on*  
5 *both concentrations and deposition of specific air pollutants. Most of the previous*  
6 *studies were about the impacts of ship emissions on global and continental scale,*  
7 *while there are only few studies available that quantify the impact of ship emissions*  
8 *on smaller scales using high resolution models. In this modeling study..*

9 [Our answers to your specific questions follow below:](#)

10 1) How are the Mozart fields translated into CAMx?

11 [We added the following text :](#)

12 *Page 30963, line 10: The initial and boundary concentrations were obtained from the*  
13 *MOZART global model for the studied period. MOZART uses geographic latitude-*  
14 *longitude coordinates and has a resolution of 1.895° x 1.875°. Data were extracted*  
15 *for the area covered by our model domain and adapted to our horizontal grid cells*  
16 *and vertical layers using our preprocessors (Oderbolz et al., 2012).*

17 2) How about biomass burning, dust and sea-salt emissions?

18 [As we mentioned in our reply to Referee # 1 who also raised the same question,](#)  
19 [emission inventory used in this study did not have wild fire, sea salt and dust](#)  
20 [emissions. Although there are some estimates of fires using the fire radiative power](#)  
21 [\(FRP\) from satellites \(Sofiev et al., 2013\), their occurrence and intensity as well as](#)  
22 [vertical distributions vary significantly spatially and temporally making their](#)  
23 [parameterization difficult. In order to avoid further uncertainties and likely errors, we](#)  
24 [decided not to include fire emissions in our simulations, until reliable data](#)  
25 [parameterization is available. On the other hand, emissions from residential heating](#)  
26 [\(wood burning\) were included in the inventory.](#)

27 [Sea salt modeling has large uncertainties mainly in generation of sea spray which](#)  
28 [occurs as the waves break on the surface of the ocean and whitecaps form \(Tsyro et](#)  
29 [al., 2011\). However, sea salt is mainly found on coarse particles and sea salt](#)  
30 [modeling would improve mainly formation of coarse nitrate \(Sellegrì et al., 2001\).](#)  
31 [Similarly, mineral dust is more relevant for coarse particles \(Athanasopoulou et al.,](#)

1 2010). Since our focus in this work was only on the fine fraction of particles ( $PM_{2.5}$ ),  
2 we believe that lack of such emissions did not affect our results significantly.

3 We added the following comments in the Methods section:

4 *page 30963, line 26: The annual emission data for 10 SNAP (Selected Nomenclature*  
5 *for sources of Air Pollution) categories per grid cell in geographic latitude–longitude*  
6 *coordinate system were converted to hourly, gridded data using the monthly, weekly*  
7 *and diurnal profiles provided by TNO. Wild fire, sea salt and mineral dust emissions*  
8 *were not included in the inventory. There are some estimates of fires using the fire*  
9 *radiative power (FRP) from satellites (Sofiev et al., 2013). Occurrence and intensity*  
10 *of such emissions as well as vertical distributions however, vary significantly spatially*  
11 *and temporally making their parameterization difficult. Sea salt is mainly found on*  
12 *coarse particles and sea salt modeling would improve mainly formation of coarse*  
13 *nitrate (Sellegri et al., 2001). Similarly, mineral dust is more relevant for coarse*  
14 *particles (Athanasopoulou et al., 2010). Since our focus in this work was only on the*  
15 *fine fraction of particles ( $PM_{2.5}$ ), we believe that lack of such emissions did not affect*  
16 *our results significantly.*

17 3) How are the anthropogenic emission distributed vertically?

18 Anthropogenic emissions are mostly treated as area emissions. If enough information  
19 about point sources is available, one can distribute such emissions to the vertical  
20 layers of the model. Some of them would be then injected to the first two layers. In  
21 this study, all emissions were treated as area emissions in the first model layer.

22 *Page 30964, line 6: All emissions were treated as area emissions in the first model*  
23 *layer.*

24 4) How are the SOA calculated (2-product, vbs, etc)? This is actually described much  
25 later in the discussions but I think it should also be described in the methodology  
26 section.

27 Calculation of SOA was described in Section 2: Method: "Calculation of secondary  
28 organic aerosols (SOA) was based on the semi-volatile equilibrium scheme called  
29 SOAP (Strader et al., 1999) that partitions condensable organic gases to seven types  
30 of secondary organic aerosols."

31 In order to clarify it we inserted the following text:

1 *This is the traditional 2-product approach which treats the primary organic aerosols*  
2 *as non-volatile.*

3 5) Figure S1 does not show the contribution of ships emissions, it shows the absolute  
4 ship emissions used in the study.

5 *This is correct and it was written as follows in page 30964, line 7 : Figure S1 shows*  
6 *the annual emissions from ships.*

7 6) How are the deposition velocities calculated?

8 *As given in Method section (page 30963, lines 14-18), dry deposition of gases in*  
9 *CAMx was calculated using a state-of-the-science, LAI (leaf-area index)-based*  
10 *resistance model (Zhang et al., 2003). This scheme possesses an updated*  
11 *representation of non-stomatal deposition pathways and has been tested extensively*  
12 *(Environ, 2011). For surface deposition of particles, CAMx includes diffusion,*  
13 *impaction and/or gravitational settling. CAMx uses separate scavenging models for*  
14 *gases and aerosols to calculate wet deposition.*

15 7) Although published, a few sentences of the model performance of the base case  
16 scenario should be written in this study.

17 *We added the following text in page 30964, line 11:*

18 *Model performance and uncertainties:*

19 *The model performance for simulations reported in this paper was thoroughly*  
20 *evaluated and the results were published in Aksoyoglu et al. (2014). It is however,*  
21 *necessary to give some information about the model performance here. Accuracy of*  
22 *the state-of-the-art air quality models such as CAMx, depends largely on the quality*  
23 *of the input data such as meteorological fields and emissions. It is well known that*  
24 *reproducing the meteorological parameters like wind fields under difficult conditions –*  
25 *especially in wintertime- is challenging. Uncertainty in emissions varies depending on*  
26 *pollutant and source. In general, some emission sources are difficult to estimate*  
27 *regionally, such as agricultural activities. For example, ammonia emissions and their*  
28 *daily and diurnal variations are related to actual climate conditions in a particular*  
29 *year. According to Kuonen et al. (2014), uncertainty estimates for emissions vary*  
30 *between 10-300% depending on pollutant and source.*

1 *Biogenic emission models require a detailed vegetation inventory, emission factors*  
2 *(based on a very few data) for each specific species as well as temperature and*  
3 *radiation data (Guenther et. al. 2012, Oderbolz et al., 2013). In spite of extensive*  
4 *efforts in this field, biogenic emission models still have high uncertainty mostly due to*  
5 *lack of sufficient measurements of these species. Evaluation of deposition is another*  
6 *challenge since measurement techniques are available only for wet deposition. Dry*  
7 *deposition can only be estimated using gas phase concentrations and dry deposition*  
8 *velocities.*

9 *By keeping these uncertainties in mind, the general performance of both WRF and*  
10 *CAMx models was reasonably good for the modeled period with some*  
11 *underestimation of  $PM_{2.5}$  during January-February when unusually high*  
12 *concentrations were reported in Europe due to severe meteorological conditions. The*  
13 *agreement between measurements and meteorological model results was good, with*  
14 *high correlation coefficients (0.76–0.98) and low mean bias error, MBE (-1.13 for air*  
15 *temperature, 0.57 for wind speed). These values fulfil the desired accuracy*  
16 *suggested by Cox et al. (1998). The model evaluation of the CAMx model suggested*  
17 *a relatively good model performance with a mean bias of 4 ppb and  $-1.9 \mu\text{g m}^{-3}$  for*  
18 *ozone and  $PM_{2.5}$  concentrations, respectively. Details of the model performance of*  
19 *the base run including ship emissions have been published in Aksoyoglu et al.*  
20 *(2014).*

21 8) Page 5, line 19: ..due to reduced NO<sub>x</sub>-titration effect by the exclusion of ships.

22 We assume that the referee means page 30964, line 22 which reads : “.. due to  
23 enhanced titration caused by NO<sub>x</sub> emissions from ships”. If this is the case, maybe  
24 this sentence needs clarification: Fig. 1 shows the difference in ozone mixing ratios  
25 between simulations with and without ships. The negative sign in the figure indicates  
26 a decrease in ozone when ship emissions are included. The base case includes ship  
27 emissions.

28 We modified the sentence in page 30964, line 6 as: *We performed CAMx simulations*  
29 *for 2006 with (base case) and without (no ship) ship emissions.*

30 We also modified all related figure captions as follows:

1 *Contribution of ship emissions (left in ppb, (base case-no ship), right in % (base*  
2 *case-no ship)x100/(base case)) to ....*

3 9) Page 7, line 21: ... of secondary aerosols produced from shipping emissions  
4 increased...

5 Unfortunately, we can't find the location of this comment since page numbers do not  
6 correspond to those in the manuscript.

7 10) I think the first paragraphs of sections 3.3.1 and 3.3.2 fits better to the  
8 introduction

9 We agree that the first paragraph in section 3.3.1 gives a short introduction about  
10 nitrogen deposition. In introduction, we tried to give some general information about  
11 the issues related to shipping emissions and their atmospheric impacts. The section  
12 3.3.1 however, is specifically about N deposition and we think that the paragraph fits  
13 better to this section.

14 On the other hand, the first paragraph in section 3.3.2 contains the results about  
15 sulfur deposition. We think therefore it should be kept in that section.

16

## 17 **References**

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1 **Contribution of ship emissions to the concentration and**  
2 **deposition of air pollutants in Europe**

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## 1 Abstract

2 Emissions from the marine transport sector are one of the least regulated anthropogenic  
3 emission sources and contribute significantly to air pollution. Although strict limits were  
4 introduced recently for the maximum sulfur content in marine fuels in the SECAs (sulfur  
5 emission control areas) and in the EU ports, sulfur emissions outside the SECAs and  
6 emissions of other components in all European maritime areas have continued to increase in  
7 the last two decades. We have used the air quality model CAMx with and without ship  
8 emissions for the year 2006 to determine the effects of international shipping on the annual as  
9 well as seasonal concentrations of ozone, primary and secondary components of PM<sub>2.5</sub> and the  
10 dry and wet deposition of nitrogen and sulfur compounds in Europe. The largest changes in  
11 pollutant concentrations due to ship emissions were predicted for summer. Concentrations of  
12 particulate sulfate increased due to ship emissions in the Mediterranean (up to 60%), in the  
13 English Channel and the North Sea (30-35%) while increases in particulate nitrate levels were  
14 found especially in the north, around the Benelux area (20%) where there were high NH<sub>3</sub>  
15 land-based emissions. Our model results showed that not only the atmospheric concentrations  
16 of pollutants are affected by ship emissions, but also depositions of nitrogen and sulfur  
17 compounds increase significantly along the shipping routes. NO<sub>x</sub> emissions from the ships  
18 especially in the English Channel and the North Sea, cause a decrease in the dry deposition of  
19 reduced nitrogen at source regions by moving it from the gas-phase to the particle phase  
20 which then contributes to an increase in the wet deposition at coastal areas with higher  
21 precipitation. In the western Mediterranean region on the other hand, model results show an  
22 increase in the deposition of oxidized nitrogen (mostly HNO<sub>3</sub>) due to the ship traffic. Dry  
23 deposition of SO<sub>2</sub> seems to be significant along the shipping routes whereas sulfate wet  
24 deposition occurs mainly along the Scandinavian and Adriatic coasts. The results presented in  
25 this paper suggest that evolution of NO<sub>x</sub> emissions from ships and land-based NH<sub>3</sub> emissions  
26 will play a significant role in the future European air quality.

27

## 28 1 Introduction

29 There have been many studies on the effects of air and road traffic emissions and projections  
30 of their future levels (Cuvelier et al., 2007; Schurmann et al., 2007; Westerdahl et al., 2008;  
31 Koffi et al., 2010; Uherek et al., 2010; Wilkerson et al., 2010; Hodnebrog et al., 2011).  
32 Relatively few studies, on the other hand, have dealt with the impacts of ship emissions in

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**Deleted:** 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.

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**Deleted:** 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.

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**Deleted:** 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.

1 detail (Corbett et al., 2007; Eyring et al., 2010; Huszar et al., 2010; Jonson et al., 2015).

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

9 Legislation on air pollutants and greenhouse gases from the maritime sector is a major  
10 challenge because of the characteristics of the shipping sector, which include global trade  
11 operations based in different countries. The efforts of the European Union (EU) and the  
12 International Maritime Organization (IMO) to tackle emissions from international shipping  
13 are different and to date there is no integrated legislation. Globally the International maritime  
14 Organization (IMO) is regulating emissions through the International Convention for the  
15 Prevention of Pollution from Ships (MARPOL) and its Annex VI,  
16 (<http://www.imo.org/OurWork/Environment/PollutionPrevention/Pages/Default.aspx>). The  
17 latest fuel sulfur limits in so-called Emission Control Areas (ECAs) were set at 0.1% as of 1  
18 January 2015. Reductions of NO<sub>x</sub> emissions from marine diesel engines are also regulated,  
19 but these focus only on new ships, where limits are defined as a function of speed and  
20 installation year.

21 In Europe, the maximum sulfur content of the marine fuel used by ships operating in the  
22 Sulfur Emission Control Areas (SECAs) - the Baltic Sea, the English Channel and the North  
23 Sea- was restricted to 1.0% in July 2010 and further reduced to 0.1% in January 2015. The  
24 EU sulfur directive has limited the sulfur content to 0.1% in harbor areas since January 2010.

25 Although more stringent NO<sub>x</sub> emission limits legislated by the International Maritime  
26 Organization (IMO) have forced marine diesel engine manufacturers to consider a variety of  
27 different emission reduction technologies, there is no NECA (NO<sub>x</sub> Emission Control Areas)  
28 in Europe yet. Since the IMO NO<sub>x</sub> emissions regulations refer only to new ships, the impact  
29 of these regulations is minimal at present and probably will continue to be so in the near  
30 future (EEA, 2013).

31 The highest level of detail on ship movements can be obtained with the AIS (Automatic  
32 Identification System) data. The AIS was developed and made compulsory by the

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1 International Maritime Organization for all ships over 300 gross tonnage to minimize the  
2 probability of groundings and collisions of ships. These signals allow very accurate  
3 positioning of vessels and their emissions. When combined with knowledge on each ship's  
4 engine and possible abatement techniques, a realistic estimation of fuel consumption and  
5 emissions can be made. Jalkanen et al. (2009) presented an automated system that is based on  
6 AIS signals, to evaluate exhaust emissions from marine traffic in the Baltic Sea area. A pilot  
7 project using the AIS data to estimate shipping emissions in the port of Rotterdam allowed for  
8 calculation of emissions on a much finer geographical grid than could be done previously  
9 (Denier and Hulskotte, 2010). In the near future, AIS data is expected to be used to improve  
10 accuracy of emission estimates in a larger area in Europe.

11 Johansson et al. (2013) reported that the emission limitations from 2009 to 2011 have had a  
12 significant effect on reducing the emissions of SO<sub>x</sub> in the northern emission control area in  
13 Europe. On the other hand, sulfur emissions in sea areas outside the SECAs and emissions of  
14 other species - especially NO<sub>x</sub> - in all sea areas around Europe have been increasing over the  
15 past decades, while land-based emissions have been gradually coming down. The revised  
16 Gothenburg Protocol specifies national emission reduction commitments in Europe to be  
17 achieved by 2020 ([http://www.unece.org/env/lrtap/multi\\_h1.html](http://www.unece.org/env/lrtap/multi_h1.html)). These commitments  
18 however are only for land-based sources and do not cover emissions from international  
19 shipping. According to the European Environment Agency, emissions of nitrogen oxides from  
20 international maritime transport in European waters are projected to increase and could be  
21 equal to land-based sources by 2020 (EEA, 2013). It is therefore important to understand the  
22 impacts of shipping emissions on both concentrations and deposition of specific air pollutants.  
23 Most of the previous studies were about the impacts of ship emissions on global and  
24 continental scale, while there are only few studies available that quantify the impact of ship  
25 emissions on smaller scales using models with high resolution. In this modeling study, we  
26 investigated the impacts of ship emissions on European air quality in detail by analyzing the  
27 seasonal and spatial variations of the contributions from the shipping sector to the  
28 concentrations of ozone and PM<sub>2.5</sub> components as well as to the deposition of nitrogen and  
29 sulfur compounds.

## 30 **2 Method**

31 The models used in this study are the Comprehensive Air quality Model with extensions,  
32 CAMx, Version 5.40 (<http://www.camx.com>) and the Weather Research & Forecasting

1 Model (WRF-ARW), Version 3.2.1 (<http://wrf-model.org/index.php>). The model domain  
2 covered all of Europe with a horizontal resolution of  $0.250^\circ \times 0.125^\circ$  which corresponds  
3 approximately to 19 km x 13 km around the central latitudes of the model domain. We used  
4 6-hour ECMWF data (<http://www.ecmwf.int/>) to provide initial and boundary conditions for  
5 the WRF model. WRF uses 31 vertical layers up to 100 hPa, of which 14 were used in CAMx  
6 with the lowest layer being 20 m thick. The initial and boundary concentrations were obtained  
7 from the MOZART global model for the studied period (Horowitz et al., 2003). MOZART  
8 uses geographic latitude-longitude coordinates and has a resolution of  $1.895^\circ \times 1.875^\circ$ . Data  
9 were extracted for the area covered by our model domain and adapted to our horizontal grid  
10 cells and vertical layers using our preprocessors (Oderbolz et al., 2012). Photolysis rates were  
11 calculated using the TUV photolysis pre-processor (<http://cprm.acd.ucar.edu/Models/TUV/>).  
12 Ozone column densities were extracted from TOMS data  
13 (<http://ozoneaq.gsfc.nasa.gov/OMIOzone.md>). Dry deposition of gases in CAMx is calculated  
14 using a state-of-the-science, LAI (leaf-area index)-based resistance model (Zhang et al.,  
15 2003). For surface deposition of particles, CAMx includes diffusion, impaction and/or  
16 gravitational settling. CAMx uses separate scavenging models for gases and aerosols to  
17 calculate wet deposition. The gas-phase mechanism used in this study was CB05 (Carbon  
18 Bond Mechanism 5) (Yarwood et al., 2005). Concentrations of particles with a diameter  
19 smaller than 2.5  $\mu\text{m}$  were calculated using the fine/coarse option of CAMx. Calculation of  
20 secondary organic aerosols (SOA) was based on the semi-volatile equilibrium scheme called  
21 SOAP (Strader et al, 1999) that partitions condensable organic gases to seven types of  
22 secondary organic aerosols. This is the traditional 2-product approach which treats the  
23 primary organic aerosols as non-volatile.

24 The gridded TNO-MACC data for 2006 were used as the basic anthropogenic emission  
25 inventory (Denier van der Gon et al., 2010). The annual emission data for 10 SNAP (Selected  
26 Nomenclature for sources of Air Pollution) categories per grid cell in geographic latitude-  
27 longitude coordinate system (with a grid resolution of  $0.125^\circ \times 0.0625^\circ$  which corresponds  
28 approximately to 9 km x 7 km around the central latitudes of the model domain) were  
29 converted to hourly, gridded data using the monthly, weekly and diurnal profiles provided by  
30 TNO. Wild fire, sea salt and mineral dust emissions were not included in the inventory. There  
31 are some estimates of fires using the fire radiative power (FRP) from satellites (Sofiev et al.,  
32 2013). Occurrence and intensity of such emissions as well as vertical distributions however,  
33 vary significantly spatially and temporally making their parameterization difficult. Sea salt is

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1 mainly found on coarse particles and sea salt modeling would improve mainly formation of  
2 coarse nitrate (Sellegrì et al., 2001). Similarly, mineral dust is more relevant for coarse  
3 particles (Athanasopoulou et al., 2010). Since our focus in this work was only on the fine  
4 fraction of particles ( $PM_{2.5}$ ), we believe that lack of such emissions did not affect our results  
5 significantly.

6 The biogenic emissions (isoprene, monoterpenes, sesquiterpenes) were calculated as  
7 described in Andreani-Aksoyoglu and Keller (1995) using the temperature and shortwave  
8 irradiance from the WRF output, the global USGS land use data and the GlobCover 2006  
9 inventory. All emissions were treated as area emissions in the first model layer. We  
10 performed CAMx simulations for 2006 with (base case) and without (no ship) ship emissions.  
11 Figure S1 shows the annual emissions from ships. Temporal profiles for ship emissions show  
12 a small increase (~10%) in summer with respect to winter (Denier van der Gon et al., 2011).  
13 Concentrations as well as dry and wet deposition of pollutants were calculated over the entire  
14 year.

15

#### 16 ***Model performance and uncertainties***

17 The model performance for simulations reported in this paper was thoroughly evaluated and  
18 the results were published in Aksoyoglu et al. (2014). It is however, necessary to give some  
19 information about the model performance here. Accuracy of the state-of-the-art air quality  
20 models such as CAMx, depends largely on the quality of the input data such as  
21 meteorological fields and emissions. It is well known that reproducing the meteorological  
22 parameters like wind fields under difficult conditions –especially in wintertime- is  
23 challenging. Uncertainty in emissions varies depending on pollutant and source. In general,  
24 some emission sources are difficult to estimate regionally, such as agricultural activities. For  
25 example, ammonia emissions and their daily and diurnal variations are related to actual  
26 climate conditions in a particular year. According to Kuonen et al. (2014), uncertainty  
27 estimates for emissions vary between 10-300% depending on pollutant and source.

28 Biogenic emission models require a detailed vegetation inventory, emission factors (based on  
29 a very few data) for each specific species as well as temperature and radiation data (Guenther  
30 et. al. 2012, Oderbolz et al., 2013). In spite of extensive efforts in this field, biogenic emission  
31 models still have high uncertainty mostly due to lack of sufficient measurements of these  
32 species. Evaluation of deposition is another challenge since measurement techniques are

1 available only for wet deposition. Dry deposition can only be estimated using gas phase  
2 concentrations and dry deposition velocities.

3 By keeping these uncertainties in mind, the general performance of both WRF and CAMx  
4 models was reasonably good for the modeled period. The model evaluation of the CAMx  
5 model suggested a relatively good model performance with a mean bias of 4 ppb and -1.9 mg  
6 m<sup>-3</sup> for annual ozone and PM<sub>2.5</sub> concentrations, respectively. There was some underestimation  
7 of PM<sub>2.5</sub> in January-February when unusually high concentrations were reported in Europe  
8 due to severe meteorological conditions. The agreement between measurements and  
9 meteorological model results was good, with high correlation coefficients (0.76–0.98) and  
10 low mean bias error, MBE (-1.13 for air temperature, 0.57 for wind speed). These values fulfil  
11 the desired accuracy suggested by Cox et al. (1998). Details of the model performance of the  
12 base run including ship emissions have been published in Aksoyoglu et al. (2014).

13

### 14 **3 Results and discussion**

#### 15 **3.1 Annual impacts**

16 The annual mean surface ozone was predicted to be about 40 ppb over the sea and coastal  
17 areas when emissions from the marine transport sector were excluded (Fig. S2). Ship  
18 emissions cause an increase in the mean surface ozone by 4-5 ppb (5-10%) in the  
19 Mediterranean Sea (Fig. 1). On the other hand, ozone levels decrease by about 5-6 ppb (10-  
20 20%) around the English Channel and the North Sea due to enhanced titration caused by NO<sub>x</sub>  
21 emissions from ships. It was shown in an earlier sensitivity study for the same year that ozone  
22 formation in that area was VOC-limited because of high NO<sub>x</sub>/VOC ratios whereas a NO<sub>x</sub>-  
23 sensitive regime was predicted for the Mediterranean region (Aksoyoglu et al., 2012).

24 The modeled mean annual concentration of PM<sub>2.5</sub> varied between 5 and 40 µg m<sup>-3</sup> for the year  
25 2006 without ship emissions in Europe (Fig. S3). The highest concentrations were predicted  
26 around the Benelux area, northern Italy and eastern Europe. The concentration of PM<sub>2.5</sub>  
27 increased along the shipping routes as well as the coastal areas when emissions from the ship  
28 traffic were included (Fig. 2). These changes were caused not only by primary PM (elemental  
29 carbon (EC) and primary organic aerosol (POA)) emissions from ships, but also by an  
30 increase in the concentration of precursor species leading to the formation of secondary PM  
31 (particulate nitrate (NO<sub>3</sub>), sulfate (SO<sub>4</sub>), ammonium (NH<sub>4</sub>) and secondary organic aerosol

1 (SOA)). The largest contribution was predicted in the western Mediterranean (up to 45%) as  
2 well as along the north European coast (10-15%). Studies with other models using the 2005  
3 inventory at a relatively coarse resolution of about 50 km showed a similar spatial distribution  
4 but predicted a lower contribution (15-25%) in the Mediterranean (EEA, 2013). The  
5 difference is probably due to the use of different emission inventories, in addition to the  
6 different domain resolutions. The finer resolution used in this study was able to capture the  
7 local effects more clearly.

## 8 **3.2 Seasonal impacts**

### 9 **3.2.1 Ozone**

10 We analyzed the changes in the surface ozone mixing ratios caused by the ship emissions in  
11 each season separately (Fig. 3). The effects were stronger in summer and there was a  
12 difference in the seasonal variation between north and south. Ship emissions were predicted to  
13 cause a decrease in ozone in the north, including the area of the English Channel, the North  
14 Sea and the Baltic Sea in all seasons except summer. Ozone decreased in summer due to ship  
15 traffic only around the English Channel by -20% while it increased by about 5% in the eastern  
16 part of the North Sea and the Baltic Sea (Fig. 3b). These results are in the same range as those  
17 found by Huszar et al. (2010) for 2004. The area around the English Channel is a high-NO<sub>x</sub>  
18 region leading to a reduction of the surface ozone concentration as a result of the contribution  
19 from ship emissions as discussed in section 3.1.

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

### 1 3.2.2 PM<sub>2.5</sub>

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

### 9 3.2.3 Impacts on aerosol components in summer

10 In this section, the contribution of ship emissions in summer to the individual components of  
11 PM<sub>2.5</sub> is investigated, because the effects are stronger in that season (see Fig. 4b). In order to  
12 understand which components are affected more by ship emissions, we first analyzed the  
13 effects on primary and secondary species. The contribution of ship emissions to the  
14 concentrations of primary and secondary PM<sub>2.5</sub> is shown in Fig. 5. Elevated concentrations of  
15 the primary carbonaceous components EC and POA were predicted only along the shipping  
16 routes in the Mediterranean and in the north around the English Channel and the North Sea  
17 (Fig. 5a) whereas the concentrations of secondary aerosols (SA) containing secondary  
18 inorganic (SIA) and secondary organic (SOA) aerosols increased over a larger area (Fig. 5b).  
19 These results suggest that the effects on the concentrations of secondary particles (via  
20 formation by oxidation of gaseous precursors) are more significant than the effects on primary  
21 particles (by direct emissions). As seen in Fig. 5b, the concentrations of secondary aerosols  
22 increased not only over the sea areas but also over the continent due to emissions from  
23 international shipping.

24 Detailed analysis of model results revealed that the change in the secondary aerosol  
25 concentration due to ship emissions occurs mainly in the inorganic fraction (Figs. 6a-c). The  
26 concentrations of particulate nitrate and ammonium increased by about 10-20% around the  
27 Benelux area and northern Italy where there are high land-based ammonia emissions (Figs. 6a  
28 and 6b). These results indicate that NO<sub>x</sub> emissions from the ships and ammonia emissions  
29 from the land lead to the formation of ammonium nitrate. On the other hand, particulate  
30 sulfate increased along the shipping routes and coastal areas with the largest effects (50-60%)  
31 in the western Mediterranean and the North African coast (Fig. 6c). The contribution to the

1 SOA concentration was relatively small (< 10%) and was mainly found in the north (Fig. 6d).  
2 We note that the results for SOA might look different if a VBS (Volatility Basis Set) scheme  
3 were used to calculate the organic aerosol (OA) concentrations (Donahue et al., 2006), but  
4 this could not be done because the volatility distribution of ship emissions is not well known  
5 yet (Pirjola et al., 2014).

### 6 **3.3 Contribution to deposition**

#### 7 **3.3.1 Nitrogen deposition**

8 The atmospheric deposition of pollutants raises serious concerns for ecosystems. In general,  
9 the main nitrogen sources are emissions of nitrogen oxides from combustion processes and  
10 ammonia from agricultural activities. The deposition of atmospheric nitrogen species  
11 constitutes a major nutrient input to the biosphere, which enhances forest growth. Despite  
12 this, increased nitrogen input into terrestrial ecosystems represents a potential threat to forests.  
13 Enhanced nitrogen deposition can cause soil acidification, eutrophication and nutrient  
14 imbalances, causing a reduction in biodiversity. The deposition of atmospheric nitrogen  
15 compounds occurs via dry and wet processes. NO<sub>2</sub>, NH<sub>3</sub>, nitric acid (HNO<sub>3</sub>), and nitrous acid  
16 (HONO) are the most important contributors to nitrogen dry deposition. Nitrogen wet  
17 deposition results from the scavenging of atmospheric N constituents.

18 The predicted annual deposition of total nitrogen in Europe based only on the land emissions  
19 varied between 5 and 45 kg N ha<sup>-1</sup> y<sup>-1</sup> in 2006 (Fig. 7, left) and it was mainly dominated by  
20 dry deposition (Fig. S4). The largest dry deposition was generally over the regions with high  
21 ambient NH<sub>3</sub> concentrations (the Benelux area and northern Italy) as also reported previously  
22 (Flechard et al., 2011). In the rest of the area, dry deposition of oxidized nitrogen dominated.

23 As seen in the right panel of Fig. 7, ship emissions caused an increase in N deposition along  
24 the shipping routes except for a few high-NH<sub>3</sub> locations where a small decrease in deposition  
25 was predicted. Analysis of the changes in the dry and wet deposition showed that the main  
26 contribution of ship emissions was to dry N deposition while wet deposition increased slightly  
27 (10%) in the North Sea (Fig. 8 and Fig. S5).

28 Further investigation of the changes in the dry deposition showed that ship emissions caused  
29 an increase in the dry deposition of HNO<sub>3</sub> in the Mediterranean whereas there was a small  
30 decrease (-2%) in the NH<sub>3</sub> deposition in ammonia-rich areas (Fig. 9). Dry deposition of  
31 ammonia occurred close to the source areas. Our results suggest that NO<sub>x</sub> emissions from

1 ships were responsible for transformation of some gaseous ammonia to particulate ammonium  
2 (see Fig. S6), which has a lower dry deposition velocity than gaseous NH<sub>3</sub> but contributes to  
3 an increased wet deposition especially over the North Sea (Fig. 8 right panel). The largest  
4 contribution of the ship traffic emissions to deposition of oxidized nitrogen (in the form of  
5 HNO<sub>3</sub>) was in the Mediterranean Sea (see Fig. 9, right panel).

### 6 3.3.2 Sulfur deposition

7 After emission, sulfur dioxide is further oxidized in the atmosphere, with sulfuric acid and  
8 sulfate as final products. Sulfate is mostly removed by wet deposition, with various effects on  
9 ecosystems including acidification of marine ecosystems and soil, vegetation damage, as well  
10 as corrosion. Excluding the ship emissions, the largest total sulfur deposition was predicted to  
11 occur in the eastern part of Europe (with high land-based SO<sub>2</sub> emissions) (Fig. 10, left) and  
12 was dominated by dry deposition (Fig. S7, left panel). Wet deposition was predicted to be  
13 relatively higher in areas with high precipitation (Fig. S7, right panel). Generally, the  
14 importance of dry deposition of sulfur decreased and the importance of wet deposition  
15 increased with distance from the source, along with the decrease of the SO<sub>2</sub> / sulfate ratio.

16 Our simulations showed that ship emissions contributed substantially to the sulfur deposition  
17 along the shipping routes and the coastal areas (Fig. 10, right panel, see Fig. S8 for relative  
18 contribution). The western Mediterranean and the North African coast were especially  
19 affected by the sulfur deposition from ship traffic. As shown in Fig. 11, the contribution to the  
20 dry SO<sub>2</sub> deposition dominated along the shipping routes while the effect on wet SO<sub>4</sub>  
21 deposition was smaller and was mostly in areas with higher precipitation. Comparison of the  
22 right panel of Fig. 10 with the left panel of Fig. 11 shows clearly that the contribution of ship  
23 emissions to sulfur deposition is mainly as SO<sub>2</sub> dry deposition.

24

## 25 4 Conclusions

26 Although regulations for emissions from the maritime traffic sector –especially for sulfur-  
27 have been tightened over the last few years, the impacts are limited at present in Europe since  
28 there is no NECA (NO<sub>x</sub> Emission Control Area) yet and the IMO emissions limits refer only  
29 to new ships. The European Environment Agency estimated that emissions of nitrogen oxides  
30 from international maritime transport in European waters could be equal to land-based  
31 sources by 2020. The model results presented in this study give an overview of the effects of

1 ship emissions on the concentrations and depositions of air pollutants in Europe, based on the  
2 2006 emission inventory.

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

16 The PM<sub>2.5</sub> concentrations increased by up to 45% in the Mediterranean Sea, and 10-15% in  
17 the North Sea, Baltic Sea and along the coastal areas due to ship traffic. The impacts predicted  
18 for the Mediterranean region are larger than those reported in other studies. The finer  
19 resolution used in this work captured the local effects more accurately. Significant effects of  
20 ship emissions on the air quality were predicted not only along the shipping routes, but also  
21 over a large part of the European continent. Although increased concentrations of primary  
22 organic aerosols and elemental carbon were predicted only along the shipping routes,  
23 secondary pollutants were affected over a larger area. The effects of ship emissions were  
24 larger in summer predominantly on secondary inorganic aerosols whereas secondary organic  
25 aerosol concentrations increased by less than 10%. One should keep in mind however, that the  
26 results for SOA might look different if a VBS (Volatility Basis Set) scheme is used to  
27 calculate the organic aerosol (OA) concentrations, but this could not be done in this study due  
28 to lack of information about the volatility distribution of ship emissions. Ship emissions  
29 increased the particulate sulfate concentrations in the Mediterranean as well as in the North  
30 Sea. On the other hand, particulate nitrate concentrations increased due to the NO<sub>x</sub> emissions  
31 from shipping, especially around the Benelux area where there are high land-based NH<sub>3</sub>  
32 emissions.

1 Consumption of gaseous  $\text{NH}_3$  for particulate nitrate formation resulted in a small decrease in  
2 the dry deposition of reduced nitrogen in its source regions and an increase in wet deposition  
3 along the shorelines with high precipitation rates. Deposition of nitrogen was predicted to  
4 increase in the Mediterranean mainly due to an increase in the deposition of oxidized nitrogen  
5 compounds (mainly  $\text{HNO}_3$ ). On the other hand, the increase in dry deposition of  $\text{SO}_2$  along  
6 the shipping routes was larger than the increase in wet deposition of  $\text{SO}_4$  along the  
7 Scandinavian and the Adriatic coast.

8 The model results achieved in this study suggest that emissions from ship traffic have  
9 significant impacts on air quality, not only along the shipping routes but also over a large part  
10 of the European continent. While  $\text{SO}_2$  emissions in European waters will continue to decrease  
11 due to regulation of the sulfur content in marine fuels,  $\text{NO}_x$  emissions are expected to increase  
12 further in the future and could be equal to or even larger than the land-based emissions from  
13 2020 onwards. Impacts of regulations for  $\text{NO}_x$  emissions from marine diesel engines are  
14 expected to be limited in the near future.

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

21 As a final remark, we have to consider the following issues for future European air quality: **in**  
22 **general, there is a clear need to improve the emission inventories to reduce the uncertainties;**  
23 **since ammonia is a very important precursor for the secondary inorganic aerosol formation,**  
24 **more accurate estimates of its emissions are needed for future simulations; with significant**  
25 **future reductions of  $\text{NO}_x$  emissions from ship traffic, changing chemical regimes around the**  
26 **northern coast would affect the impacts on ozone as well as the formation of secondary**  
27 **inorganic aerosols.** Decreasing  $\text{NO}_x/\text{VOC}$  ratios would affect ozone formation whereas  
28 decreasing  $\text{NO}_x/\text{NH}_3$  ratios might change the formation of secondary inorganic aerosols as  
29 well as nitrogen deposition since ammonia land emissions are not expected to decrease  
30 significantly in the near future compared to sulfur and nitrogen emissions in Europe.

31

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11

12

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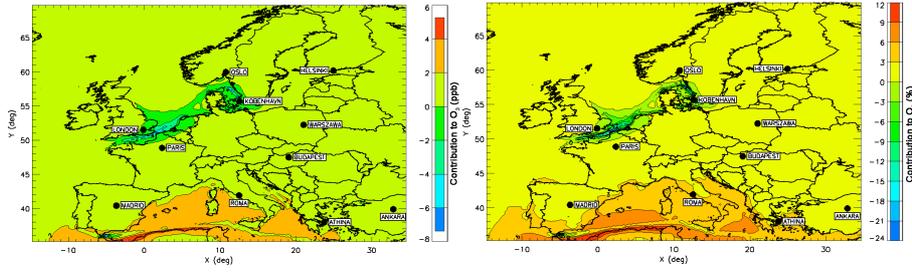
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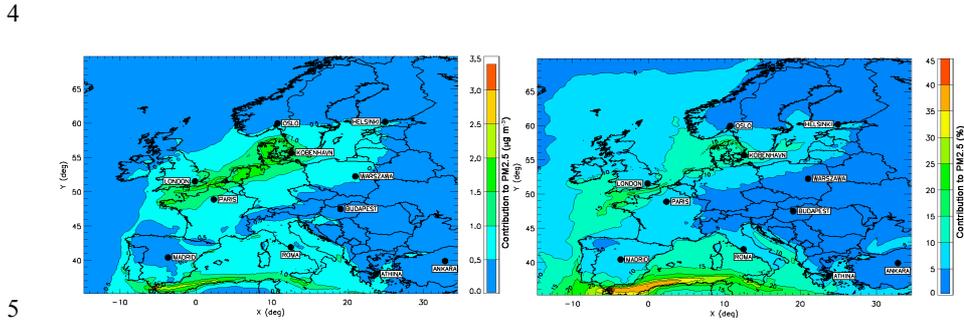
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1  
 2 Figure 1. Contribution of ship emissions to mean surface O<sub>3</sub> in 2006: left in ppb (base case-no  
 3 ship), right in % ((base case-no ship)x100/(base case)).



5  
 6 Figure 2. Contribution of ship emissions to the mean PM<sub>2.5</sub> concentration in 2006: left in µg  
 7 m<sup>-3</sup> (base case-no ship), right in % ((base case-no ship)x100/(base case)).

8

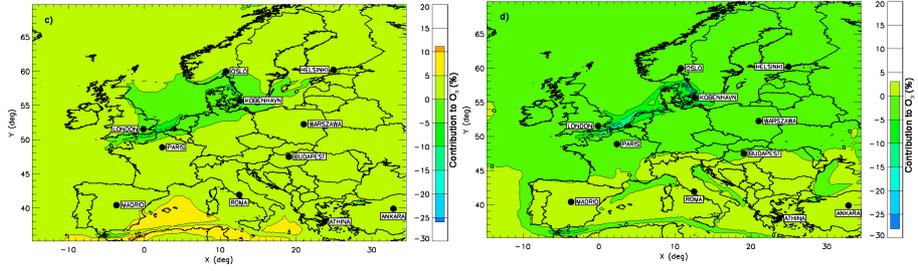
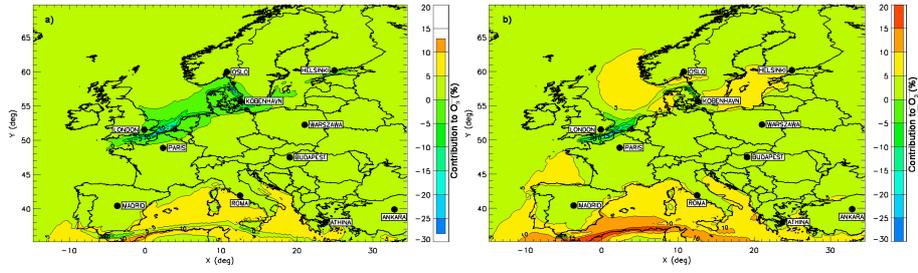
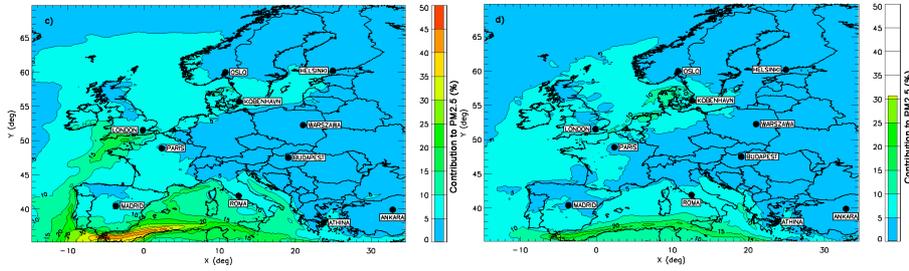
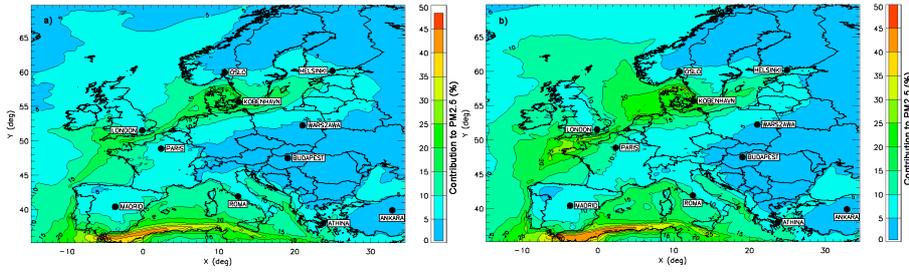
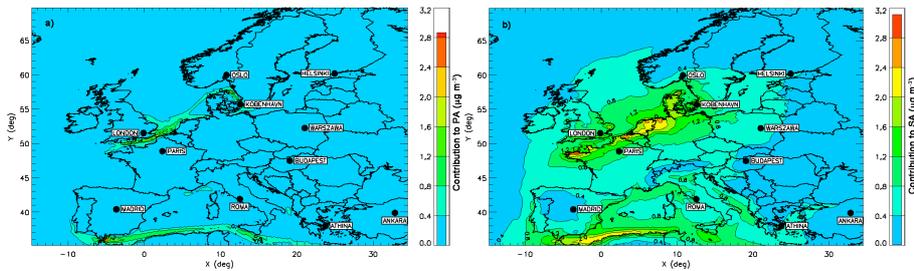


Figure 3. Contribution of ship emissions to mean surface O<sub>3</sub> (%) in a) spring, b) summer, c) fall, and d) winter  $((\text{base case-no ship}) \times 100 / (\text{base case}))$ .

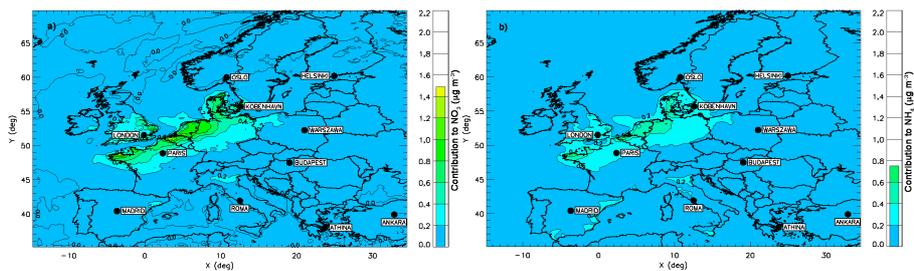


1  
2  
3 Figure 4. Contribution of ship emissions to PM<sub>2.5</sub> (%) in a) spring, b) summer, c) fall, and d)  
4 winter  $((\text{base case-no ship}) \times 100 / (\text{base case}))$ .

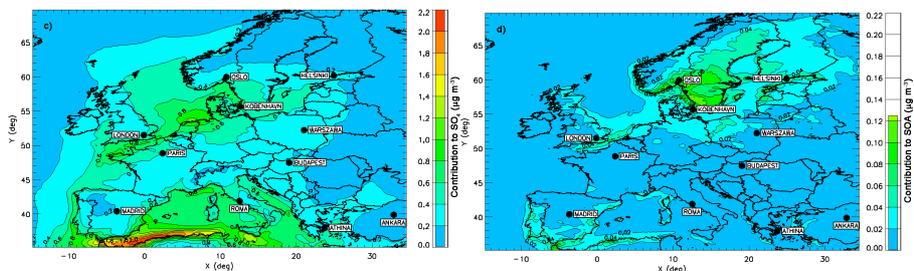
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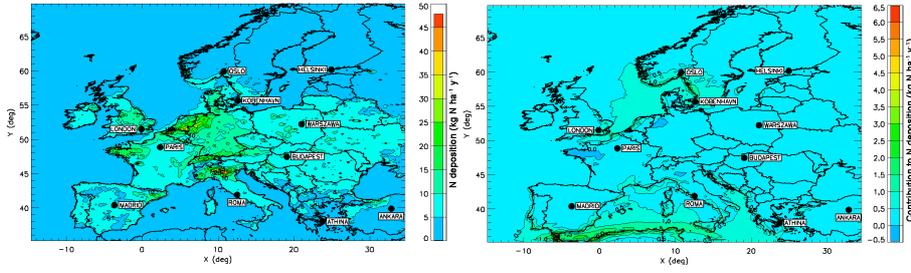


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 2 Figure 5. Contribution of ship emissions ( $\mu\text{g m}^{-3}$ ) to a) the primary aerosol (PA) b) the  
 3 secondary aerosol (SA) concentration in summer 2006 (base case-no ship).

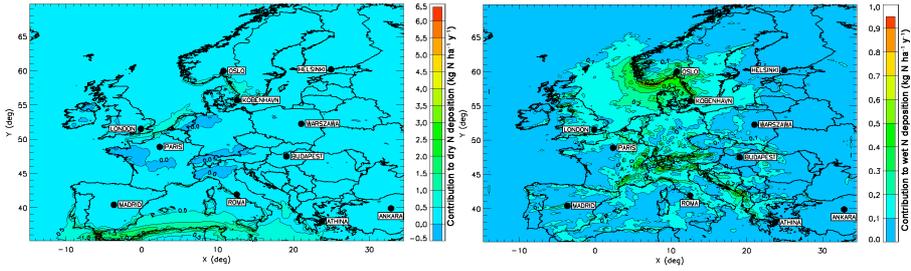


5  
 6  
 7 Figure 6. Contribution of ship emissions ( $\mu\text{g m}^{-3}$ ) to the secondary aerosol concentration; a)  
 8  $\text{NO}_3$  b)  $\text{NH}_4$  c)  $\text{SO}_4$  d) SOA in summer 2006 (base case-no ship). Note that the scale in (d) is  
 9 ten times smaller than the others.

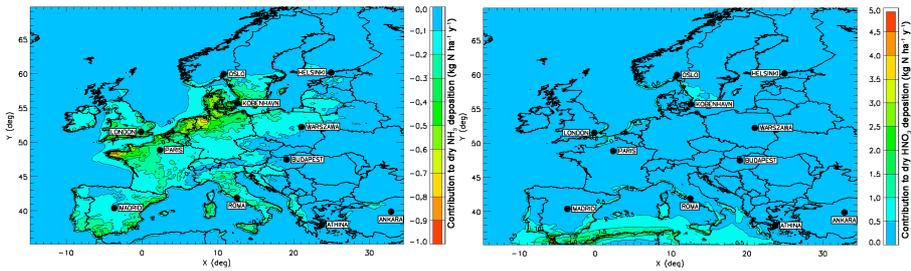




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 2 Figure 7. Annual nitrogen deposition only due to land-based emissions (left) and contribution  
 3 of ship emissions to N deposition (right) (base case-no ship).

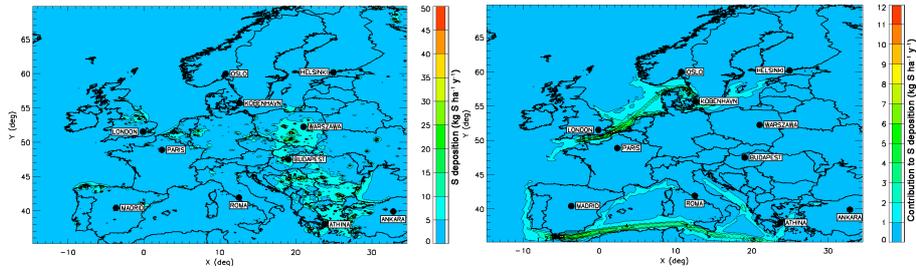


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 6 Figure 8. Contribution of ship emissions to the annual dry N deposition (left) and wet N  
 7 deposition (right) (base case-no ship).

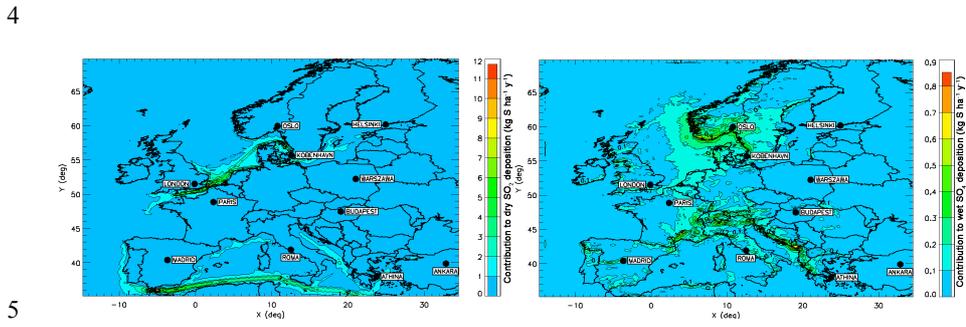


9  
 10 Figure 9. Contribution of ship emissions to the annual dry NH<sub>3</sub> deposition (left) and dry  
 11 HNO<sub>3</sub> deposition (right) (base case-no ship).

12



1  
 2 Figure 10. Sulfur deposition only due to land-based emissions (left) (no ship) and due to ship  
 3 emissions (right) (base case-no ship).



5  
 6 Figure 11. Contribution of ship emissions to dry SO<sub>2</sub> deposition (left) and wet SO<sub>4</sub> deposition  
 7 (right) (base case-no ship).

8  
 9