Reply to comments of Anonymous Referee # 2:

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

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

Thank you for your comments and suggestions. In order to clarify the motivation of this study we will add the following text in the introduction:

Page 30962, line 11: Although more stringent NOx emission limits legislated by the International Maritime Organization (IMO) have forced marine diesel engine manufacturers to consider a variety of different emission reduction technologies, there is no NECA (NOx Emission Control Areas) in Europe yet. Since the IMO NOx emissions regulations refer only to new ships, the impact of these regulations is minimal at present and probably will continue to be so in the near future (EEA, 2013).

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

Our answers to your specific questions follow below:

1) How are the Mozart fields translated into CAMx?

We will add the following text :

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

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

As we mentioned in our reply to Referee # 1 who also raised the same question, emission inventory used in this study did not have wild fire, sea salt and dust emissions. Although there are some estimates of fires using the fire radiative power (FRP) from satellites (Sofiev et al., 2013), their occurrence and intensity as well as vertical distributions vary significantly spatially and temporally making their parameterization difficult. In order to avoid further

uncertainties and likely errors, we decided not to include fire emissions in our simulations, until reliable data parameterization is available. On the other hand, emissions from residential heating (wood burning) were included in the inventory.

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

We will add the following comments in the Methods section:

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

3) How are the anthropogenic emission distributed vertically?

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

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

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

Calculation of SOA was described in Section 2: Method: “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.”

In order to clarify it we will amend the following text:

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

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

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

6) How are the deposition velocities calculated?

As given in Method section (page 30963, lines 14-18), dry deposition of gases in CAMx was calculated using a state-of-the-science, LAI (leaf-area index)-based resistance model (Zhang et al., 2003). This scheme possesses an updated representation of non-stomatal deposition pathways and has been tested extensively (Environ, 2011). 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.

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

We will add the following text in page 30964, line11:

Model performance and uncertainties:

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

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

By keeping these uncertainties in mind, the general 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. The agreement between measurements and meteorological model results was good, with high correlation coefficients (0.76–0.98) and low mean bias error, MBE (-1.13 for air temperature, 0.57 for wind speed). These values fulfil the desired accuracy suggested by Cox et al. (1998). The model evaluation of the CAMx model suggested a relatively good model performance with a mean bias of 4 ppb and -1.9 $\mu g\ m^{-3}$ for ozone and $PM_{2.5}$ concentrations, respectively. Details of the model performance of the base run including ship emissions have been published in Aksoyoglu et al. (2014).

8) Page 5, line 19: ..due to reduced NOx-titration effect by the exclusion of ships.

We assume that the referee means page 30964, line 22 which reads : ".. due to enhanced titration caused by NOx emissions from ships". If this is the case, maybe this sentence needs clarification: Fig. 1 shows the difference in ozone mixing ratios between simulations with and without ships. The negative sign in the figure indicates a decrease in ozone when ship emissions are included. The base case includes ship emissions.

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

We will then modify all related figure captions as follows:

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

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

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

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

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

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

References

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