Reply to comments of Anonymous Referee # 1:

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

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

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

Major comments

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

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

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

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

The model performance for simulations reported in this paper was thoroughly evaluated and the results were published in Aksoyoglu et al. (2014). 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. As shown in Aksoyoglu et al. (2014), results of meteorological model WRF for this application were quite satisfactory.

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

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.

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

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

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).

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

We will add the following in page 30971, line 4:

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

We will add the following in page 30972, line 1:

As a final remark, we have to consider the following issues for future European air quality: in general, there is a clear need to improve the emission inventories to reduce the uncertainties; since ammonia is a very important precursor for the secondary inorganic aerosol formation, more accurate estimates of its emissions are needed for future simulations; with significant future reductions of NOx 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.

The authors describe their methods, regarding the MACC and biogenic emissions. However, they should also clearly state, which emission categories were NOT include- that is good scientific practice. As MACC includes only anthropogenic emissions, they probably neglected at least wild fire, sea salt and dust emissions. If all of these were neglected, they should at least provide

some estimate (using proper references) on how large a fraction of emissions for each relevant pollutant was not taken into account. The neglected source categories have a direct influence on the contribution percentages of shipping, compared with total concentrations and depositions.

The emission inventory used in this study did not have wild fire, sea salt and dust emissions. There are some estimates of fires using the fire radiative power (FRP) from MODIS equipped 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. 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.

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.

It should also be reported what was the spatial resolution of the emission inventory (in kilometers), especially regarding the shipping emissions. The authors should also report the resolution of their chemical transport modelling (CTM) not only in terms of degrees; but for readability, also report what these correspond as kilometers in the domain used.

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

Page 30963, line 6: The model domain covered all of Europe with a horizontal resolution of 0.250^{o} x 0.125^{o} which corresponds approximately to 19 km x 13 km around the central latitudes of the model domain.

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 (with a grid resolution of 0.125^{o} x 0.0625^{o} which corresponds approximately to 9 km x 7 km around the central latitudes of the model domain) were converted to hourly, gridded data using the monthly, weekly and diurnal profiles provided by TNO.

Minor comments

Abstract. “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 former part of this sentence is vague (‘differently’, not stated in which respect), and the latter part is trivial. Remove or clarify the former part, and delete the latter.

“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.” Trivial statement, to be removed.

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.” Trivial statements, to be removed.

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

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

Line 9. WRF occurs once too many

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

Thank you for these comments. We will revise them accordingly.

References

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