AUTHOR'S RESPONSES TO REFEREE #1:

We thank Referee #1 for positive evaluation and for taking the time to read and give valuable comments to improve the manuscript. Following the reviewer remarks, we addressed the comments and questions in detail.

1)"Model validation: no evidence is preported that the model was validated with observations. Was it validated in any way? How? Even if it cannot be validated with actual shipping contributions due to the different methods used (lines 237-238), the authors could compare their results to total NO2 or other gaseous pollutant concentrations from reference stations, for example. This kind of comparison would be essential to confirm their modelling results."

Answer: Thank you for your comments. Although information about model validation can be found in lines 129-134: "Regarding the performance of the model, simulations from EMEP/MSC-W are regularly evaluated against measurements in the EMEP annual reports (Norwegian Meteorological Institute, 2018). Moreover, there are several studies that compare model results with measurements and calculations with other models (Angelbratt et al., 2011; Bessagnet et al., 2016; Colette et al., 2011, 2012; Jonson et al., 2010; Karl et al., 2017; Prank et al., 2016; Soares et al., 2016) and recent studies that used the model to assess the effects of shipping emissions (Jonson et al., 2015, 2017; Turner et al., 2017)", in order to support our results, model output $PM_{2.5}$, PM_{10} and NO_2 concentrations for the S-SCN scenario were compared with data from the monitoring stations of EU Member States reported by the European Environmental Agency for 2015. Moreover, comparisons between the modelling reference results reported by EMEP for the year 2015 were also compared with the data from the monitoring stations. Annual mean concentrations observed in 139 stations for $PM_{2.5}$, 337 stations for PM_{10} and 446 stations for NO_2 were compared with the model results in time and space. Information about model validation will be added in the Methods section as follows: "…and recent studies that used the model to assess the effects of shipping emissions (Jonson et al., 2015, 2017; Turner et al., 2017). To support the results of the present study, model output $PM_{2.5}$, PM_{10} and NO_2 concentrations for the S-SCN scenario were compared with data from the monitoring stations of EU Member States reported by the European Environmental Agency for 2015 (EEA, 2020). Moreover, comparisons between the modelling reference results reported by EMEP for the year 2015 (Norwegian Meteorological Institute, 2019) were also compared with the data from the monitoring stations. Annual mean concentrations observed in 139 stations for $PM_{2.5}$, 337 stations for PM_{10} and 446 stations for NO_2 were compared with the model results in time and space. Table 1 summarizes the model quality indicators (Pearson correlation coefficient (Pearson's r), Mean Bias Error (MBE), Mean Absolute Error (MAE) and Root Mean Square Error (RMSE)), for the present study and for the reference results reported by EMEP. Similar results were obtained for

the comparison with the present study and with the reference results of EMEP, which indicates that the model simulations were well executed. Correlations obtained were moderately positive (Pearson's r > 0.5) for all pollutants, with errors smaller than those reported in the literature (Monteiro et al., 2018).”

Table 1. Model quality indicator values for the present study and for the reference results reported by EMEP.

Indicators	This study			EMEP reference		
	$PM_{2.5}$	PM_{10}	NO_2	$PM_{2.5}$	PM_{10}	NO_2
Pearson's r	0.57	0.55	0.70	0.64	0.55	0.67
MBE [a]	1.32	19.51	5.78	0.34	18.70	5.19
MAE [b]	2.86	19.55	8.70	2.81	18.74	9.18
RMSE [c]	3.62	20.83	11.24	3.59	20.11	11.90

[a] Mean Bias Error; [b] Mean Absolute Error; [c] Root Mean Square Error

2) Primary vs. secondary aerosol contributions: it is unclear in the manuscript whether the particle concentrations modelled are primary or primary + secondary aerossol from shipping. If secondary aerosols were included, how was this implemented in the model? This is the main limitation of most models targeting shipping emissions (both dispersion and receptor models). Please address this carefully in the Methods section.

Answer: Thank you for your comments. The secondary aerosols were included in the model. In the EMEP MSC-W model $PM_{2.5}$ concentrations were defined as $PM_{2.5} = SO_4^{2-} + NO_3^-$(fine) + NH_4^+ + SS(fine) + $PPM_{2.5}$ + 0.27 NO_3^-(coarse) considering the secondary organic aerosols as the aerosol mass arising from the oxidation products of gas-phase species, the secondary inorganic aerosols as $SO_4^{2-} + NO_3^-$(fine) + NH_4^+ + NO_3^-(coarse), sea salt (SS) and the primary particulate matter ($PPM_{2.5}$ and PPMcoarse) originating directly from anthropogenic emissions (as was the case of shipping emissions). PM_{10} concentrations were calculated as $PM_{10} = PM_{2.5}$+PMcoarse where PMcoarse was defined as PMcoarse = 0.33 NO_3^-(coarse) + SS(coarse) + PPMcoarse. Information about how PM concentrations were modelled in this study will be added in the Methods section as follows: “… having a thickness of 50 m. PM concentrations were modelled considering primary particulate matter originating directly from anthropogenic emissions, as well as secondary organic and inorganic aerosols and sea salt. Other details about the model can be found in Simpson et al. (2012) and in Norwegian Meteorological Institute (2017a).”

Specific comments:

- line 25, " its contribution", does this refer to health impacts? The contribution to air quality degradation has been assessed in numerous papers in the literature, including the papers referenced by the authors.

Answer: Yes, we were referring to the contribution for human health degradation. We decided to change to: "… which may lead to known negative effects on air quality and health, being its contribution to human health degradation still not well documented (Brandt et al., 2013; Corbett et al., 2007; Nunes et al., 2017b; Sofiev et al., 2018)."

- the English could be reviewed by a native speaker, it is good but some small typos remain.

Answer: Suggestion attended. The manuscript will be review by a native speaker.

- line 33, suggestion to reference the EEA report EEA, 2013. The impact of international shipping on European air quality and climate forcing. EEA Technical Report 04/2013. Luxembourg: Publications Office of the European Union, 2013. ISBN 978-92-9213-357-3.

Answer: Suggestion attended. The reference will be added.

- Please add in the Methods section discussions on model validation and on secondary aerosols (whether they are or not included in the model).

Answer: Suggestion attended. More information about model validation will be added, according to our previous answers.

- line 150, what does "ash" refer to, exactly? Please define

Answer: Thanks for your comment. Once in STEAM PM emissions were calculated as the sum of SO4, H2O, EC, OC and ash, considering the different emission factors, we chose to maintain this separation. Ash refers to a component of the PM emitted by ships and depends on the content of marine fuels. To give more information about the ash component and emission factors used in STEAM we will add the following sentence: "… sulphates and ash (a component of the PM emitted by ships that depends on the content of marine fuels) for the Iberian Peninsula in 2015 in a 0.1ºx0.1º grid cells (approximately 10 x 10 km^2). Details about emission factors used in STEAM can be found in Jalkanen et al. (2009), Jalkanen et al. (2012) and Jonson et al. (2014)."

- line 155, "ports", the resolution is quite coarse (10x10 km2) to represent harbour emissions, or even most coastal urban areas. Please highlight this as a limitation.

Answer: Thanks for your comment. Our objective was not to make a detailed analysis of emissions or concentrations in ports. Despite the limitation of the grid used (10x10km), it was possible to identify higher emissions for the cells near the port areas. Anyway, we will add the fact that this resolution is too coarse to make a detailed analysis of emissions and concentrations in ports as a limitation of the study in the "Uncertainties and limitations"

section as follows: "…Furthermore, EMEP-MSC/W model has been recently compared with the CMAQ and the SILAM models and showed the best spatial correlation of annual mean concentrations for NO2, SO2 and PM2.5 resulting of shipping emissions, although it seems to be underestimating PM2.5 concentrations and overestimating O3 concentrations. Moreover, although it has been possible to identify variations in the emissions and concentrations near the port areas, the resolution that was used was too coarse to make a detailed analysis of emissions and concentrations inside the port areas."

- line 160, suggest to check and reference the report HEI Special report 22, Impacts of shipping on air pollution emissions, air quality, and health in the Yangtze River Delta and Shanghai, China

Answer: Thank you for your comment. We checked the results of the report HEI Special report 22 and information about the differences in the emissions intensities will be added as follows: "Nevertheless, in the HEI report authors described lower emission intensities for the Yangtze River Delta and Shanghai areas at 12 NM from the coast. According to these results, comparisons should be made carefully as emission intensities seem strongly dependent on the location for which they are calculated (inside the port area, at a certain distance from the coast or on the high seas) and also on the methodology used to calculating shipping emissions."

- lines 185-188, please add a statistical trend analysis: the differences don't seem statistically significant, to the naked eye.

Answer: Thank you for your comment. Statistical trend analysis will be added. The ranked non-parametric test Mann-Kendall trend test was used for detecting monotonic trends in the monthly emissions. The null hypothesis H0 was assumed as "there is no trend in the emissions over the months" and it was tested against the alternative hypothesis H1 which considered that "there is increasing or decreasing trend in the monthly emissions". The tests performed at the 95% confidence interval level showed no statistically significant trends in the monthly emissions data. Information about statistical trend analysis will be added as following: "It can be observed that emissions increased progressively from February to July, where they reached the maximum annual value. After that, a decrease during August and September was observed, followed by a stabilization during October (for some pollutants there was a slight increase) and a decrease until December. Although emissions varied throughout the year, variations were about 1-2% between months and each month represented 7.1-9.1% of the annual total emissions. In fact, according to the statistical trend analysis using the Mann-Kendall trend test, performed at the 95% confidence interval level, no statistically significant variations were achieved in the monthly emissions data for all pollutants (p-values > 0.05)."

- line 191: I don't think the comparison with a paper from 1999 (even if a reference paper) is adequate here: in 20 years the trade and sailing patterns have surely changed largely, therefore this comparison is not representative.

Answer: Suggestion attended. This information will be deleted from the manuscript.

- line 203, are these primary or secondary PM10 and PM2.5 concentrations? Or the sum of both?

Answer: These are primary and secondary PM_{10} and $PM_{2.5}$. Details about this issue were already described in a previous answer.

- line 206, why do concentrations increase gradually towards and over the N of Africa? Are there no O3 sinks (e.g., major cities) in this region? This seems unlikely, probably the emission inventories are not accurate for this region. Please discuss.

Answer: Thanks for your comment. Actually, it is not over the North of Africa but close to it, near the coast, thus on the sea area, and not over the region. That is why there are no major cities there.

- line 217, are these (4.8 microg/me and 6.9 microg/m3) shipping contributions? They seem quite high, especially if only primary aerosols are considered (I'm still unsure of this). Also, are these average values for the entire peninsula? Please compare with shipping contributions from the literature, and also with total (non-ship sourced) PM10, NO2, O3, etc concentrations.

Answer: Thank you for your comments. As already mentioned primary and secondary aerosols were considered for modelled PM concentrations. These values are not average values, but maximum values that were verified in one grid cell of the domain. Comparisons with average values were performed with other studies and are in lines 220-224.

- line 240-241: these contributions seem unlikely as they are reported here. What distance to the coast do these results refer to? Even in coastal areas shipping is seldom the main contributing source, almost always surpassed by traffic contributions (see for Spain the works by Pandolfi et al., Pérez et al, iana et al., Amato et al....). It seems unlikely that shipping accounts for 50% of NO2 ambient concentrations inland. Or are the authors referring to air emissions? If so, this could be possible for major cities such as Barcelona. Please clarify the meaning of these sentences.

Answer: Thank you for your comments. These results are referring to the contribution of ship emissions to annual mean concentrations calculated as [((S-SCN) – (B-SCN)) / (B-SCN)] x 100. Moreover, these results (the 50% contribution) refer to inland zones close to the biggest port areas (as can be seen from Figure 4 a)). It was also possible to identify contribution of around 75% for inland regions close to the Strait of Gibraltar.

- line 249: once again, model validation is needed here.

Answer: Thank you for your comment. Model validation will be added as follows: “Monteiro et al. (2018) reported for the west coast of Portugal (also the west coast of Iberian Peninsula) lower contributions for NO_2 and PM_{10} (higher than 20% and less than 5%, respectively) than those reported in this study probably due to the different methodology applied. Moreover, according to the model validation made by Monteiro et al. (2018), their model underestimated PM_{10} and NO_2 concentrations (negative MBE), while the model used in the present study overestimated them (positive MBE).”

- lines 265-268 and 283-284: please remove the references to the "port of", as the model’s resolution is too coarse to capture this.

Answer: Thank you for your comment. We will change to “area close of Port”.

- section 3.3, please add model validation and the issue of primary and secondary aerosols, as limitations.

Answer: Thank you for your comment. Model validation will be added as above described. Primary and secondary aerosols were considered in the model, thus this is not a limitation of the study.

- line 333, suggestion to add reference to the HEI Special Report

Answer: Suggestion attended. The reference will be added.