Dear Editor,

Please fin attached the point-by-point response to the reviews.

Thank you.

Kind regards,

Sofia Sousa

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.

AUTHOR'S RESPONSES TO REFEREE #2:

We thank Referee #2 for the positive evaluation and for taking the time to read our paper and giving us valuable comments to improve the manuscript. Following the reviewer remarks, we addressed the comments and questions in detail below.

1)"The study evaluates the impacts of shipping emissions on the air quality in the region of the Iberian Peninsula and the Strait of Gibraltar, one of the busiest maritime routes in the world. This chemistry-transport modelling study makes use of shipping emissions generated by the STEAM 3 model that allocates ship activities via the Automatic Identification System operating on board the vessels. Among the valuable information presented in this manuscript are a comparison of ship emission intensities with those reported for ports in the Asian region and a calculation of the ship impact on exceedances of regulatory air quality limits. Unfortunately, it is not immediately apparent what the manuscript adds to already published chemistry transport modelling studies on the impact of ship emissions in Europe. Overall, the manuscript reads more like a good technical report than a research article, as the applied methods are not originally proposed and the uncertainties of model results are not comprehensively discussed and quantified."

Answer: Thank you for your comments. With this study, we try to give an overall view of the air quality impact of the shipping emissions over the Iberian Peninsula (specifically) using the STEAM, that is considered one of the most reliable methods to estimate shipping emissions (exhibits the highest spatially resolution in their emissions and a large number of secondary routes that do not appear in other inventories), and EMEP for the air quality modelling. Only another study estimated the concentrations for the Iberian Peninsula, but used different methodologies to do it (different inventory and CTM, referred in lines 69-78).

Furthermore, this is the first time that the modelling results of this new version of STEAM are discussed in detail for the Iberian Peninsula (this inventory was already discussed but from a global point of view, not specifically for the Iberian Peninsula) and the very first time that they were used to estimate the pollutant concentrations for the Iberian Peninsula.

Moreover, to support our results a model validation will be added. 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 the 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. Also, a comparison between the exceedances for the scenario including ship emissions and the levels from the monitoring stations was also made. We estimate the percentage of exceedances that our model found in relation to the exceedances detected with the concentrations of the monitoring stations. More information was added to the Uncertainties chapter to improve the discussion.

2) "The spatial resolution is the same as in prior studies that covered the whole of Europe. This is somewhat surprising, given that a prior study by Monteiro et al. (2018) in the same region used a finer resolution (3 km x 3km). The applied difference method for quantifying the shipping emission contribution is flawed since the effect of nonlinearities in ozone chemistry on the ship impact was not evaluated, despite the high photochemical activity in this region. For both daytime and night time, the instantaneous NOx lifetime in ship plumes is a strong function of the initial NOx concentration at ship stack, resulting in a very nonlinear loss rate for NOx in ship plumes (e.g. Song et al.,2003). Model procedures that shift ship plume levels by an order of magnitude, as can be expected for a 10-km wide grid cell, will quite likely overestimate NOx lifetime."

Answer: Thank you for your comments. As shipping is a major source of NOx to the troposphere, and especially because large amounts of these pollutants are often released from point sources into the relatively clean maritime atmosphere, since the version rv4.8 of the EMEP MSC-W a new pseudo-species "ShipNOx" has been introduced and NOx released by ships started to be treated differently and not like any other source of NOx. Like you said in your comment, in 3-D models NOx emissions are typically diluted into large grid volumes which can lead to large over-predictions in O3 production, and in the NOx lifetime. Tests with the EMEP model confirmed these issues and also that the early approach was not appropriate for the European area at least (considered ship-related NOx emissions like any other source of NOx). To prevent such effects, the model assigns 50% of shipping NO_x to the pseudo-species ShipNOx and the rest as NO and NO_2 as previously done. ShipNOx deposits as NO_2, but suffer simple atmospheric reactions:

ShipNOx + OH⇒HNO3 Reaction 1; ShipNOx⇒HNO3 Reaction 2

Reaction 1 proceeds with the same rate as the normal NO2+ OH reaction, thus proceeding faster in daylight and in areas with high-OH. Reaction 2 provides a minimum half-life of about 6 hours, accordingly to Vinken et al. (2011) results.

The heat release from ship stack exhaust of large ships represents a buoyancy flux that may result in plume rise. Therefore, we can expect that a significant fraction of the shipping emissions are emitted at upper heights. The STEAM 3 model should be able to take into account plume rise of ship exhaust in generalized form. A

description of the treatment of the vertical distribution of shipping emissions and injection heights that are used for the corresponding vertical layers of the EMEP modelling system should be added to the method section. When shipping emissions have been fully transferred to the lowest vertical model layer, such a procedure has to be justified and the error due to this needs to be approximated.

Answer: Thank you for your comments. The estimated height distribution of emissions from STEAM are given in the figure below. According to these estimates, over 80% of emissions occur between 30-60 m height. The plume release height is a function of vessel type, size and plume rise. For the two former, vessel stack height is determined from photographs and existing data from IHS Markit. However, the airdraft (how high the vessel is from the water surface) or the keel-to-mast height are rarely available in commercially available databases. For this reason, we have used vessel scale drawings and photographs from Significant Ships publication serie, to link vessel types and sizes to stack height. There is a linear dependency between vessel stack height and vessel length, but these linear functions are vessel type specific. Regardless, STEAM does not consider plume rise, because exhaust temperature, exhaust velocity and funnel pipe diameter are not known. In principle, some typical values could be used, but currently STEAM only adds a constant value of 10m to stack height estimation to provide a primitive estimate of the plume rise.

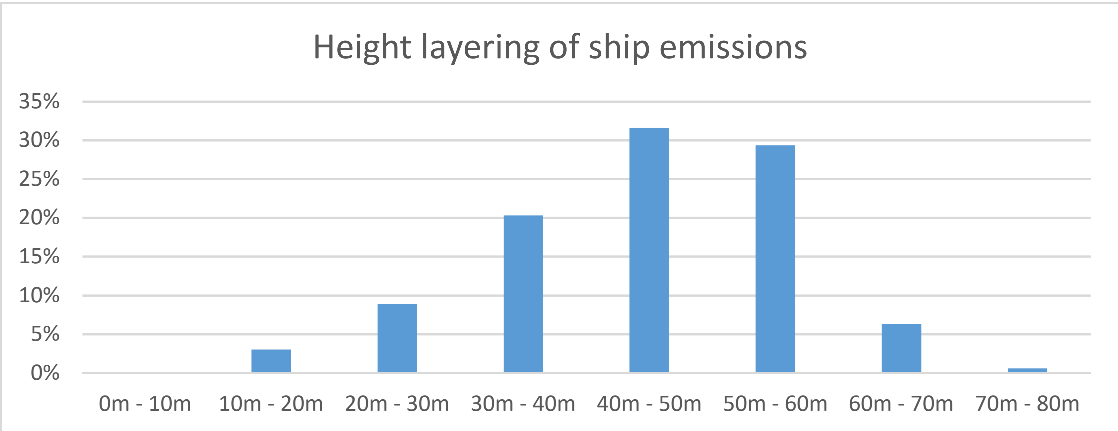

Figure 1 Estimated height profile for ship emissions around the Iberian Peninsula domain.

The detailed height profile for emissions obviously depends on the type of traffic operating in the area. Largest cruise vessels can have very high stacks, exceeding 70m height and plume rise may elevate the plumes even higher. However, STEAM does not currently consider meteorology during emission calculation. In our opinion, the plume rise issue in ship exhaust dispersion is most relevant for local scale air quality assessments, but less so for regional scale work. For this reason, the ship emissions from STEAM were allocated to the first model layer of the EMEP runs. Information related to this issue will be added to the Methods section as follows: "… having a thickness of 50 m. Assuming that the plume rise issue in ship exhaust dispersion is more relevant for local scale air quality assessments, and less for regional scale work. For this reason, the ship emissions from STEAM were allocated to the first model layer of the EMEP runs."

"The significance of the modelled ship contribution was not validated with measurements. Although the Norwegian Meteorological Institute regularly validates the air quality predictions with the EMEP MSC-W model for Europe, it is not sufficient to simply refer to this. The manuscript should include a validation of the modelled concentrations in the subdomain region with monitoring data from stations in Portugal, Spain and France for 2015 (EMEP network, EEA AirBase, EBAS database). The comparison should include model data from both runs with and without shipping emissions."

Answer: Thank you for your comments. 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 the 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 the 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 estimations and for the reference results reported by EMEP. Similar quality indicators were obtained for the comparison the results of the present study and the reference results of EMEP, which indicates that the model simulations were well executed. Although the correlations obtained were moderate positive correlations (Pearson's r > 0.5) for all pollutants, the errors obtained were smaller than those reported in the literature (Monteiro et al., 2018), which makes our results acceptable."

Table 1. Model quality indicators for the present study estimations 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

Specific Comments:

1.) P.1 lines 24-26: Many studies can be found about the impacts of shipping emissions on air quality and health. It would be a good place here to discuss deviations and contradictions in the literature concerning the relevance of shipping for health impacts, and specifically the roles of primary versus secondary particulate matter.

Answer: Suggestion attended. We will change the following sentences: "Marine traffic has been identified as a relevant source of pollutants especially nitrogen oxides (NO_x), sulphur oxides (SO_x) and particulate matter (PM), 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). Studies have been reporting that shipping contributions to ambient PM in port areas are mainly secondary particles (around 60 to 70% of PM_{10} and $PM_{2.5}$ mass concentrations). Despite this, studies have been suggesting that could be more advantageous to reduce shipping-related primary particle emissions than precursors of secondary particles (NO_x and SO_x), which are the target of current international regulations (Viana et al., 2014)."

2.) P.4 lines 111-114: Which boundary conditions of the chemical concentrations were used for the subdomain runs?

Answer: Information about boundary conditions used in the chemical transport model for the subdomain runs will be added as: "…400 km from the Iberian Peninsula coast. The initial and the lateral boundary conditions for most of the chemical compounds were defined by functions defining concentrations in terms of latitude and time, based on measurements and/or model calculations, providing robustness which chemical transport model results sometimes lack. More information about the EMEP/MSC-W configuration for initial and boundary concentrations used in this study can be found in Simpson et al. (2012)."

3.) P. 5 line 157 to P. 6 line 173: Suggest to transfer the information of annual average emission intensities (per pollutant and per port/sea area) into a table to facilitate the comparison with shipping activity in the Asian region.

Answer: Suggestion attended. We will change the following sentences: "The annual average intensities of ash, CO, CO_2, EC, NO_x, OC, sulphate and SO_x emissions were 9.0E-04 tonnes/yr/km^2, 1.38E-02 tonnes/yr/km^2, 8.47 tonnes/yr/km^2, 1.27E-01 tonnes/yr/km^2, 1.97E-01 tonnes/yr/km^2, 3.16E-03 tonnes/yr/km^2, 8.04E-03 tonnes/yr/km^2 and 1.01E-01 tonnes/yr/km^2, respectively. The annual average and highest intensities for NO_x and SO_x reported for the Asian Region are present in Table 3 (Chen et al., 2016a, 2017; Fan et al., 2016). In general, the average intensities that were reported for Asia were considerably higher than those found in this study. It was possible to identify in the present study two main hubs given the high emissions intensity: Valencia Port and the Strait of Gibraltar. At Valencia Port, ash, CO, EC and OC had the highest values, respectively, 1.46E-01 tonnes/yr/km^2, 1.85 tonnes/yr/km^2, 1.99E-01 tonnes/yr/km^2 and 5.09E-01 tonnes/yr/km^2. At the Strait of Gibraltar, CO_2, NO_x, sulphate and SO_x had the highest values, respectively, 1330 tonnes/yr/km^2, 24 tonnes/yr/km^2, 1.03 tonnes/yr/km^2 and 11.6 tonnes/yr/km^2. In accordance to what was referred above, in the Asian Region maxima intensities were also higher than those here estimated (Chen et al., 2016b; Fan et al., 2016; Ng et al., 2013)." We will also add Table 3.

Table 3. Annual average and highest intensities of NO_x and SO_x (in tonnes/yr/km^2) reported from researches in Asian Region.

Study	Port/sea area	NO_x		SO_x	
		Annual average	Highest value	Annual average	Highest value
Chen et al. (2016a)	Tianjin Port	5.06	1.51E+03	7.14	1.79E+03
Chen et al. (2017)	Qingdao Port	1.83	-	1.42	-
Fan et al. (2016)	East China Sea	1.0	1.0E+04	1.90	1.30E+03
Ng et al. (2013)	Hong Kong	-	1.1E+02		2.0E+02

4.) Impact on Air Quality: Suggest to divide section 3.2 in topical subsections; for example "Annual average concentrations" (P. 7 lines 202 - 218), "Comparison with previous studies in the region" (P. 7 line 219 to P. 8 line 250), "Seasonal variation"

(P.8 lines 251 - 260), "Possible health impacts" (P. 8 line 261 to P.9 line 297). Some passages could be shortened.

Answer: Suggestion attended. We will subdivide the section 3.2.

5.) P. 9 lines 276-287: Suggest to illustrate the contribution of shipping emissions to the exceedances of limit values in form of a bar diagram, i.e. showing the increment of number of exceedances (NO2, PM2.5, PM10) and number of days of exceedances(SO2) due to ship traffic for the major ports of the Iberian Peninsula.

Answer: Thank you for your comment. We believe that with a bar diagram the spatial distribution (illustrated with Figure A2) will be lost.

6.) Uncertainties and limitations: The uncertainties of the emission factors of pollutants from different ship types could easily dominate the uncertainty of the evaluated contribution from shipping. With the STEAM 3 model at hand, it should be possible to estimate the overall uncertainty in the modelled concentrations due to uncertain emission factors. To arrive at a more reliable margin of the contribution of shipping emissions in this region, my request to the authors is that they perform shipping emission calculations with the respective lower and upper bound of the emission factors of NOx, SOx and primary particulates, then repeating the runs with EMEP MSC-W using the lower and higher emission dataset.

Answer: Thank you for your comments. In STEAM, there are several sources of uncertainty which can have an impact on the accuracy of the results. These could be classified in three categories:

a) Gaps in input data (incomplete AIS coverage, missing IHS Markit data)
b) Power prediction (weather contributions, Hollenbach resistance inaccuracy, fouling, squat, sea currents, aux engine power profiles, engine load estimation, power transmission, propeller properties)
c) Emission factors (specific fuel oil consumption, fuel type, fuel sulphur content allocation, engine generation)

Each of these three categories can have multiple contributions, indicated by various error sources in parenthesis. STEAM has mechanisms to mitigate most of the uncertainties listed above and some are features, like weather, are currently developed, but will be reported separately at a later stage. Uncertainties concerning emission factors may be larger for products of incomplete combustion, like CO, NMVOC, OC and EC, than for CO_2, or NO_x, because these are strongly related to engine load, engine generation and service history. The emission factors may also depend on the fuel type assignment and fuel sulphur content, which are estimated based on engine properties and maximum sulphur content allowed in each region at the time period of the study. However, the emission factors for incomplete combustion products may be affected by engine service history and thus are notoriously difficult to estimate. We are not currently aware of any study which would provide uncertainty evaluations for all emission sectors and emission factors included in regional air quality modelling and it seems curious to us to demand one for shipping, only. To conduct such a study would require many computer simulations and significant additional effort. Even if these tests would be limited to uncertainty evaluations of three air pollutants modelled by STEAM, it would still require low- and high-bound runs with STEAM and consecutive analysis with the EMEP model. Even then, the uncertainty evaluation would not be just about emission factors, because primary particulates would also require adjusting the assumptions concerning how the fuel sulphur content was assigned and what are its consequences on PM components. This will multiply the work required by at least a factor of six, which is not currently possible due to limitations in available research funding.

Technical Corrections:

P. 1 lines 17-18: "ktonnes y-1" is not a SI unit.

Answer: Given that the values are high, it is usual to present the results in above referred units. We maintained the units to be more coherent with the literature.

P. 1 line 27: on a global scale?

Answer: Suggestion attended. Yes, it is on a global scale. This information will be added.

P. 1 lines29-30: reference(s) for this statement missing.

Answer: Suggestion attended. The reference will be added.

P. 3 lines 66-67: suggest to reference the study of Ramacher et al. (2019) on local scale for Baltic Sea ports.

Answer: Suggestion attended. The reference will be added.

P. 5 line 150: "ash" – what is this chemically? Please define.

Answer: Suggestion attended. "Ash" will be defined.

P. 8 line 232: please replace "lower increases contributions" by "lower positive contributions".

Answer: Suggestion attended.

Conclusions: the word "verify" is used several times in the conclusions section (P. 10,line 321; P. 10, line 324; P. 11, line 330). Verification implies the comparison of model results to the true values, which are not known. Please change wording.

Answer: Suggestion attended. The word “verify” will be changed by “observe” and “detect”.

P. 11 line 340: what about the code availability of STEAM 3? Please include a statement here.

Answer: Suggestion attended. The following statement will be added:” STEAM model is intellectual property of the Finnish Meteorological Institute and is not publicly available”.

P. 15 lines 469-471: the citation of Marelle et al. is incomplete.

Answer: Suggestion attended. The reference will be changed.

Table 1 and Table 2: “tonne y-1” is not SI unit.

Answer: Given that the values are high, it is usual to present the results in above referred units. We maintained the units to be more coherent with the literature.

Figure 1 and Figure 2: please use SI units in labels, axis annotations and captions.

Answer: Given that the values are high, it is usual to present the results in above referred units. We maintained the units to be more coherent with the literature.

Figure 4f: what is the cause for high O3 values along the North African coast overwater?

Answer: Thank you for your comment. There are no O3 sinks in this region.

AUTHOR'S RESPONSES TO REFEREE #3:

We thank Referee #3 for the positive evaluation and for taking the time to read our paper and giving us valuable comments to improve the manuscript. Following the reviewer remarks, we addressed the comments and questions in detail below:

Major comment

My main criticism of the manuscript is that the results on additional exceedances of air quality standards due to ship emissions (which are potentially of interest to policymakers) depend strongly on the quality of the modelled fields. Only if they give a good representation of the actual air quality and exceedances, then the difference between the results with and without ship emissions can be trusted. I therefore believe that the authors need to include a comparison of the modelled concentrations and exceedances for the scenario including ship emissions to those measured by in-situ air quality net-works to demonstrate that they are close enough to reality to make interpretation of delta exceedances worthwhile.

Answer: Thank you for your comments. 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 the 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 the 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 estimations and the reference results reported by EMEP. Similar quality indicators were obtained for the comparison of the results of the present study and the reference results of EMEP, which indicates that the model simulations were well executed. Although the correlations obtained were moderate positive correlations (Pearson's r > 0.5) for all pollutants, the errors obtained were smaller than those reported in the literature (Monteiro et al., 2018), which make our results acceptable."

Table 1. Model quality indicators for the present study estimations and 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

Moreover, a comparison between the exceedances for the modelled scenario including ship emissions and those calculated with the data from the monitoring stations was also made. We were able to compare only the exceedances for $PM_{2.5}$, PM_{10} and NO_2 since we didn't have daily SO_2 concentrations data from the monitoring stations. Information about the comparison of the exceedances found with the model and with the data from the stations will be added in the Methods section as follows: "…and PM_{10} (20 μg m^{-3} for annual mean) (European Comission, 2018; WHO, 2018). To support the results of the present study, $PM_{2.5}$, PM_{10} and NO_2 exceedances found for the S-SCN scenario were compared with those calculated with data from the monitoring stations of the EU Member States (EEA, 2020). For $PM_{2.5}$, the exceedances to the WHO guideline found with the modelled data represented more than 60% of the exceedances calculated with the data from the stations. Regarding PM_{10}, a small agreement was found, with only 11% of the exceedances found for the modelled data. However, for NO_2 all the exceedances were estimated with the modelled data. According to these results, the model seems to predict with good reliability the exceedances of $PM_{2.5}$ and NO_2. For PM_{10} the results need to be used with caution."

Minor comments

While the manuscript is overall well written, it would benefit from proof reading by a native speaker.

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

page 4, line 120: Are Sahara dust emissions and NOx from lightning really taken from the NCAR fire inventory?

Answer: No. Only the forest fire emissions were taken from the NCAR fire inventory. To improve the comprehension of the sentence we decided to delete the part of "from the Fire INventory from NCAR version 1.5" and keep the reference of the inventory and add the Simpson et al. (2012) reference where more information about these emissions can be found.

page 7, line 204 and figures: I think it is stated nowhere that when you talk about concentrations, that always means at the surface (I assume)

Answer: Thank you for your comment. The concentrations are surface concentrations. Modifications will be introduced in the line referred and the figure legend.

page 8, line 251 and following: I'm a bit confused by this discussion of the origins of the seasonality. It sounds as if it is not really clear what the origin is, but don't you have all the information on the magnitude of emissions from STEAM so that you can give clear answers on what drives the seasonality?

Answer: Thank you for your comment. This analysis was related to the concentrations. Although we had all the information on the magnitude of emissions from STEAM, there are other factors that can influence the concentrations over the seasons. To understand if statistically significant differences in concentrations between the various seasons exist, the non-parametric test Kruskal-Wallis test was used to compare multiple samples (the four seasons) and the non-parametric Wilcoxon signed-rank test was used to compare related samples (two by two). Moreover, as our previous analyses claim it was during spring and summer that were registered the highest emission amounts. Information about the statistical analyses above referred will be added in the Methods section as follows: "The non-parametric test Kruskal-Wallis for multiple samples (the four seasons) and the non-parametric Wilcoxon signed-rank test for two by two samples analyses performed at the 95% confidence interval level were used to detect statistically significant variations for all pollutants in the seasonal concentration data." Moreover, we will change the Results section as follows: "Regarding the seasonal concentration data, statistically significant variations were found for all pollutants across all seasons (p-values < 0.05). In fact, according to the model results, the higher contributions of shipping emissions to the concentrations levels were registered during spring and summer periods (warm season). This pattern seems to be related to the increase in ship traffic during summer due to better meteorological conditions that allow better navigation conditions, which increases the traffic and subsequently the emissions and atmospheric pollution. Moreover, during summer months, the number of passenger ships tends to increase (due to recreational travel), especially in the Mediterranean Sea, which led to an increase of shipping emissions and their contributions to the pollutant's concentration levels."

page 10, line 299: The discussion on uncertainties and limitations is very general indeed and mainly lists the obvious. I think that the comparison to real data will make this section also more relevant.

Answer: Thank you for your comment. Comparison with real data will be added according to a previous comment.

Figure 1: I'm not sure that it makes really sense to show all these figures here – they all look the same with the colour scale chosen and I do not see what I can learn from 8 figures which I cannot already see in the first.

Answer: Thank you for your comment. In a previous version of the manuscript, we had the same scale for all pollutants which allowed to see the differences in the quantities of each pollutant. We will change it back to that version.

Shipping emissions in the Iberian Peninsula and its impacts on air quality

Rafael A.O. Nunes[1], Maria C.M. Alvim-Ferraz[1], Fernando G. Martins[1], Fátima Calderay-Cayetano[2], Vanessa Durán-Grados[2], Juan Moreno-Gutiérrez[2], Jukka-Pekka Jalkanen[3], Hanna Hannuniemi[3], Sofia I.V. Sousa[1]

[1]LEPABE – Laboratory for Process Engineering, Environment, Biotechnology and Energy, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465, Porto, Portugal
[2]Departamento de Máquinas y Motores Térmicos, Escuela de Ingenierías Marina, Náutica y Radioelectrónica, Campus de Excelencia Internacional del Mar (CEIMAR), Universidad de Cádiz, Spain
[3]Finnish Meterological Institute, P.O. Box 503, 00101 Helsinki, Finland

Correspondence to: Sofia I.V. Sousa (sofia.sousa@fe.up.pt)

Abstract. Marine traffic has been identified as a relevant source of pollutants, which cause known negative effects on air quality. The Iberian Peninsula is a central point in the connection of shipping traffic between the Americas and Africa and the rest of Europe. To estimate the effects of shipping emissions inland and around the Iberian Peninsula, EMEP MSC-W model was run considering and not considering shipping emissions (obtained with STEAM3 model). Total estimated emissions of CO, CO_2, SO_x, NO_x and particulate matter (subdivided in elementary carbon (EC), organic carbon (OC), sulphate and ash) for the study domain in 2015 were, respectively, 49 ktonnes y^{-1}, 30000 ktonnes y^{-1}, 360 ktonnes y^{-1}, 710 ktonnes y^{-1}, 4.5 ktonnes y^{-1}, 11 ktonnes y^{-1}, 32 ktonnes y^{-1} and 3.3 ktonnes y^{-1}. Shipping emissions increased SO_2 and NO_2 concentrations especially near port areas and also increased the O_3, sulphate, and particulate matter ($PM_{2.5}$ and PM_{10}) concentrations around all over the Iberian Peninsula coastline (especially in the south coastal region). Shipping emissions were responsible for exceedances of WHO air quality guideline for $PM_{2.5}$ in areas far from the coastline, which confirms that shipping emissions can contribute negatively to air quality, both in coastal and in inland areas.

1 Introduction

Marine traffic has been identified as a relevant source of pollutants especially nitrogen oxides (NO_x), sulphur oxides (SO_x) and particulate matter (PM), 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). Studies have been reporting that shipping contributions to ambient PM in port areas are mainly secondary particles (around 60 to 70% of PM_{10} and $PM_{2.5}$ mass concentrations). Despite this, studies have been suggesting that could be more advantageous to reduce shipping-related primary particle emissions than precursors of secondary particles (NO_x and SO_x), which are the target of current international regulations (Viana et al., 2014). In fact, international shipping represents around 13% and 12% of total global anthropogenic emissions of NO_x and SO_x, respectively (IMO, 2015). Moreover, according to Klimont et al.

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(2017), PM emissions from international shipping contribute with about 3–4 % to global emissions, which is comparable to the contribution of road transport. As far as known, it is up to 400 km from the coast that 70% of ship emissions occur (Eyring et al., 2009). As pollutants can be transported hundreds of kilometres towards the mainland, ships may contribute to air quality degradation in coastal areas, as well as in inland areas (Corbett et al., 2007; Eyring et al., 2009). Over the past 10 years, interest has been growing in studying the impact on air quality of maritime emissions in cities and ports using experimental measures (Contini et al., 2011; Merico et al., 2016, 2017; Pandolfi et al., 2011; Viana et al., 2015; Wang et al., 2019) and applying air quality models (AQMs) at local, regional and global levels (Abrutytė et al., 2014; Aksoyoglu et al., 2016; Aulinger et al., 2016; Barregard et al., 2019; Chen et al., 2017, 2018; EEA, 2013; Eyring et al., 2007; Lauer et al., 2007; Liu et al., 2017; Marelle et al., 2016; Marmer and Langmann, 2005; Matthias et al., 2016; Monteiro et al., 2018; Sotiropoulou and Tagaris, 2017). Nevertheless, the use of AQMs, such as CMAQ, WRF, CAMx, EMEP MSC-W and others entails inevitable sources of uncertainties and some limitations, mostly conditioned by the resolution of the models, the methodological limitations as a result of the complexity of air quality assessment, the quality of the meteorological data and, the reliability of emissions inventories (Karl et al., 2019). In the last years, the activity-based method using the Automatic Identification System (AIS) has been commonly accepted as the most accurate way to estimate shipping emissions, based on the detailed information of ship specifications and the operational data. Several authors have applied this methodology, although estimations with the Ship Traffic Emission Assessment Model (STEAM) have been recognized as the best way of conducting a reliable ship emissions inventory based on ship activity (Aulinger et al., 2016; Marmer and Langmann, 2005; Nunes et al., 2017b; Russo et al., 2018). Aulinger et al. (2016) recognized in their study that the STEAM model could be more reliable than other methods using AIS to describe ship movements. Marelle et al. (2016) evaluated emissions estimated with STEAM2 and compared them with airborne measurements from the ACCESS (Arctic Climate Change, Economy and Society) aircraft campaign. They concluded that the use of STEAM2 lead to reasonable predictions of NO_x, SO_2, and O_3 in comparison with ACCESS profiles. In addition, in a study performed by Nunes et al. (2017a) that reviewed studies from 2010 based on activity-based methodology to estimate shipping emissions, STEAM model was indicated as the best procedure to predict ships power, leading to better predictions of ships movements and more reliable emission calculations. Additionally, Russo et al. (2018) reviewed and compared five different European inventories (EMEP, TNO-MACC_III, E-PRTR, EDGAR and STEAM) including or calculating emissions from shipping; this study concluded that STEAM inventory should be used for studies requiring high-resolution shipping emissions data. STEAM allows assessing emissions from each individual ship, combining highly detailed AIS data and technical knowledge of the ships (characteristics and operative mode). STEAM is currently on its third version. From the first to the second version, carbon monoxide (CO) and particulate matter (PM) emissions were included. The method of analysing ships resistance on the water was revised and modelling of the power consumption of auxiliary engines was improved. In the third version, improvements include methods to compensate the lack of technical information of some ships and satellite data in some regions, as well as, some refinements, allowing to account legislative regulations (emission control areas, on-board emission abatement equipment and fuel sulphur content) (Jalkanen et al., 2012; Johansson et al., 2017). The majority of the studies on the impact of shipping emissions on air quality was performed for global scales (Dalsøren et al., 2009; Eyring et al., 2007; Lauer et al., 2007), using OsloCTM2 , CMAQ and ECHAM5/MESSy1-MADE models; continental scales were also addressed (Aksoyoglu et al., 2016; Marelle et al., 2016; Ramacher et al., 2019; Sotiropoulou and Tagaris, 2017), especially the Asian region (Chen et al., 2018; Liu et al., 2017; Zhang et al., 2017), using models with coarser resolutions (CAMx, WRF/Chem, CMAQ and GISS-E2 global models). There are only a few studies based on modelling results that considered the impacts of shipping emissions in local scale (Abrutytė et al., 2014; Aulinger et al., 2016; Matthias et al., 2016; Monteiro et al., 2018; Vutukuru and Dabdub, 2008) . Moreover, only few have used STEAM to estimate shipping emissions, namely for the North Sea (Aulinger et al., 2016; Jonson et al., 2015), Baltic Sea (Barregard et al., 2019; Jonson et al., 2015) and northern Norway region (Marelle et al., 2016). As far as known, there is only one local study that considered specifically the Iberian Peninsula domain, evaluating the impact of maritime emissions on air quality at European and national scales using the WRF-CHIMERE modelling system for 2016 (Monteiro et al., 2018), but not using STEAM. Shipping emissions in that study were extracted from TNO-MACC_III inventory, a high spatially resolved anthropogenic emissions data source available for Europe. This inventory has a high spatially resolution data, and the MACC-III version is an updated version with a new trend analysis for emissions for international shipping, but STEAM exhibits the highest spatially resolution in their emissions and a large number of secondary routes that do not appear in the former inventory, making emissions predicted with STEAM more precise. It also includes the disruptive changes in environmental regulations (Emission Control Areas, EU Sulphur directive) concerning sulphur in marine fuels. Moreover, it was highlighted by the MACC-III project team a clearly necessity of more research for getting data of shipping emissions (van der Gon et al., 2017; Russo et al., 2018).

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The Iberian Peninsula is the most western point of the European continent and the only natural opening by sea between the Mediterranean and the Atlantic Ocean. Considering the strategical position of the Iberian Peninsula regarding international maritime transport and the need of reducing the above referred scientific gaps, this study aimed to: i) estimate shipping emissions based on STEAM3 for 2015; ii) quantify the impacts of shipping emissions on the ambient air quality of the Iberian Peninsula using the EMEP/MSC-W model; and iii) investigate the inland regions where the European Commission air quality standards and WHO air quality guidelines were exceeded due to shipping.

2 Methodology and materials

2.1 Study area

The Iberian Peninsula is located in the southwest of Europe, mainly constituted by Portugal and Spain territories (also includes Andorra and Gibraltar). It is bordered on the southeast by the Mediterranean Sea (coastline with ≈ 1 600 km), and on the north, west, and southwest by the Atlantic Ocean (coastline with ≈ 1 650 km) being a central point in the connection of shipping traffic between the Americas and Africa and the rest of Europe (Global Ocean Associates, 2004a, 2004b, 2004c). In fact, the Iberian Peninsula has a central position between the English Channel and the Strait of Gibraltar, which are two busiest maritime routes in the world (Columbia University Press, 2001a, 2001b). Fig: A1 shows shipping traffic lines for 2015 that shows the relevance of the Iberian Peninsula in the international shipping traffic context.

2.2 Shipping emissions inventory

The shipping emissions inventory for the Iberian Peninsula in 2015 was obtained from a full bottom-up approach, using STEAM. This model combines: i) the shipping activity information from the terrestrial and satellite-based Automatic Identification System (AIS) and the technical characteristics of each ship (from HIS Markit); ii) the engine type for over ninety-thousand ships and; iii) the emission factors for each type of ship and size, engine type and mode of operation to calculate emissions from each ship. According to the above information, STEAM allows calculating the power consumptions and loads of each engine, as well as the quantity of fuel consumed to overcome a specific speed based on the resistance of each ship (Jalkanen et al., 2009). The model also permits to calculate shipping emissions as a function of time and location (Jalkanen et al., 2012; Johansson et al., 2013, 2017). Emissions of CO, CO_2, SO_x, NO_x and particulate matter (subdivided in EC, OC, sulphates and ash) were estimated for the Iberian Peninsula, from ships with an IMO number (ships for which it is mandatory using AIS equipment) and some small vessels for which the IMO number is not mandatory but with a Mobile Maritime Service Identity (MMSI) that produced a valid response during 2015. To compare shipping emissions with land-based emissions, the sum of the annual mean emissions of NO_x and SO_x from the other 11SNAP sectors for the domain of this study were calculated. Shipping emissions were analysed for monthly and seasonal patterns. Seasonal patterns were based on data from: i) January, February and March called as "winter"; ii) April, May and June called as "spring"; iii) July, August and September called as "summer"; and iv) October, November and December called as "autumn". The non-parametric test Kruskal-Wallis for multiple samples (the four seasons) and the non-parametric Wilcoxon signed-rank test for two by two samples analyses performed at the 95% confidence interval level were used to detect statistically significant variations for all pollutants in the seasonal concentration data.

2.3 EMEP modelling system - configuration and evaluation

The open-source EMEP/MSC-W chemistry transport model, version rv4.15 was used to evaluate the contribution of shipping emissions to NO_2, SO_2, $PM_{2.5}$, PM_{10}, sulphate and O_3 concentrations in the Iberian Peninsula. Model was run on a subdomain that extends from -14.25ºE to 9.05ºE and 32.15ºN to 47.35ºN, and concentrations were simulated up to approximately 400 km from the Iberian Peninsula coast. The initial and the lateral boundary conditions for most of the chemical compounds were defined by functions defining concentrations in terms of latitude and time, based on measurements and/or model calculations, providing robustness which chemical transport model results sometimes lack. More information about the EMEP/MSC-W configuration for initial and boundary concentrations used in this study can be found in Simpson et al. (2012). The model was designed for two scenarios: i) shipping scenario (S-SCN) considering shipping emissions and ii) baseline scenario (B-SCN) not considering shipping emissions. Runs were made for 2015 with a horizontal resolution of 0.1°x0.1° (long-lat) and an hourly data output. Emissions (for the same year of the shipping emissions inventory) from other sources such as, industry, road traffic, public power and among other sectors, split in 11 SNAPs, were obtained from the European emission inventories that are reported under the LRTAP Convention and the NEC Directive (EMEP/CEIP, 2018). Emissions from shipping sector considered in the inventory were excluded to avoid double counting of emissions. Moreover, it was also considered the emissions of the dust from Sahara, NOx from lightning and from forest fires (Simpson et al., 2012; Wiedinmyer et al., 2011). The model is divided into 34 vertical layers with the lowest layer having a thickness of 50 m. Assuming that the plume rise issue in ship exhaust dispersion is more relevant for local scale air quality assessments, and less for regional scale work. For this reason, the ship emissions from STEAM were allocated to the first model layer of the EMEP runs. .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). The meteorological data for 2015 were generated by the European Centre for Medium-Range Weather forecasts with the Integrated Forecast System model. According to EMEP Status Report 1/2017, 2015 was among the warmest years in Europe with temperatures reported above normal in winter and extremely high during summer in Southern Europe. Despite this, in the Iberian Peninsula temperatures below average were registered due to a persistent south-westerly flow (Norwegian Meteorological Institute, 2017b). In spring, a prolonged high pressure was established over the Iberian Peninsula leading to above-average temperatures in Portugal and Spain. In July, Spain was affected by an extraordinary and long-lasting heatwave (Norwegian Meteorological Institute, 2017b). 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). 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). The annual mean concentrations for each inland grid cell were compared with reference standards and guidelines (WHO and EU), aiming to evaluate exceedances and/or non-compliances of NO_2, SO_2, $PM_{2.5}$ and PM_{10} due to shipping emissions. Comparisons were performed considering the international reference values for pollutants in ambient air namely: i) EU air quality standards for NO_2 (40 µg m^{-3} for annual mean), SO_2 (125 µg m^{-3} for daily mean), $PM_{2.5}$ (25 µg m^{-3} for annual mean) and PM_{10} (40 µg m^{-3} for annual mean); and ii) WHO air quality guidelines for NO_2 (40 µg m^{-3} for annual mean), SO_2 (20 µg m^{-3} for daily mean), O_3 (SOMO35 - yearly sum

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of the daily maximum of 8 h running average over 35 ppb in ppb per days), $PM_{2.5}$ (10 µg m^{-3} for annual mean) and PM_{10} (20 µg m^{-3} for annual mean) (European Comission, 2018; WHO, 2018). To support the results of the present study, $PM_{2.5}$, PM_{10} and NO_2 exceedances found for the S-SCN scenario were compared with those calculated with data from the monitoring stations of the EU Member States (EEA, 2020). For $PM_{2.5}$, the exceedances to the WHO guideline found with the modelled data represented more than 60% of the exceedances calculated with the data from the stations. Regarding PM_{10}, a small agreement was found, with only 11% of the exceedances found for the modelled data. However, for NO_2 all the exceedances were estimated with the modelled data. According to these results, the model seems to predict with good reliability the exceedances of $PM_{2.5}$ and NO_2. For PM_{10} the results need to be used with caution.

3 Results and Discussion

3.1 Shipping emissions – spatial and seasonal variation

Table 2 summarizes the amount of emitted air pollutants from shipping and from land-based anthropogenic sources. Comparing NO_x and SO_x, shipping emissions with land-based emissions, on average the first were lower than the latter. Despite this, if NO_x and SO_x shipping emissions were added to the land-based emissions, the total would increase by 45% and 62%, respectively. Moreover, compared with emissions from the SNAP of road transport (660 ktonnes y^{-1} of NOx and 7.1 ktonnes y^{-1} of SOx), the emitted amounts of NO_x and SO_x from shipping were 1.1 and 51.3 times higher, respectively. These results show the importance of shipping emissions for these two pollutants.

Fig: 1 shows the annual mean shipping emissions of CO, CO_2, SO_x, NO_x, EC, OC, sulphates and ash (a component of the PM emitted by ships and 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. (2015). As can be seen, the spatial distribution was similar for all pollutants. In general, the highest emissions were established along the west coast of the Iberian Peninsula (including all Portuguese coast), in the Strait of Gibraltar and in the Mediterranean Sea, especially close to the African coast, which is consistent with world shipping traffic density (Fig: A1). It is important to emphasise that the grid cells along the coast where ports are located had also higher emissions due to hotelling activities. Although emissions during hotelling only represent a slight part of the total shipping emissions, port areas are significant receptors of these emissions due to the concentration of ships for long periods of time in some cases (Nunes et al., 2017a). The annual average and highest intensities for NO_x and SO_x reported from researches in Asian Region are present in Table 3 (Chen et al., 2016a, 2017; Fan et al., 2016). In general, the average intensities that were reported for the Asia were considerably higher than those found in this study. It was possible to identify in the present study two main hubs given the high emissions intensity: Valencia Port and the Strait of Gibraltar. At Valencia Port, ash, CO, EC and OC had the highest values, respectively, 1.46E-01 tonnes/yr/km^2, 1.85 tonnes/yr/km^2, 1.99E-01 tonnes/yr/km^2 and 5.09E-01 tonnes/yr/km^2. At the Strait of Gibraltar, CO_2, NO_x, sulphate and SO_x had the highest values, respectively, 1330 tonnes/yr/km^2, 24 tonnes/yr/km^2, 1.03 tonnes/yr/km^2 and 11.6 tonnes/yr/km^2. In accordance to what was referred above, in the Asian Region maxima intensities were also higher than those here estimated (Chen et al., 2016b; Fan et al., 2016; Ng et al., 2013). The big differences between the average and highest emission intensities of the present study and those of the Asian studies, appear to be related with the high intensity and type of maritime traffic and to the restrict fuel regulations in Europe. In fact, seven of the ten largest container ports in the world are located in China, and Asia is the region with the highest world seaborne trade, characterized by high traffic of container ships that have already been documented as one of the most pollutant category of ships (Chen et al., 2018; Ng et al., 2013; Nunes et al., 2017b; Song and Shon, 2014; UNCTAD, 2017). Moreover, since 2010, a 0.1% maximum sulphur requirement for fuels was stablished for ships at berth in EU ports, however, in China, there are only some domestic emission control areas with 0.5 % maximum (Reuters, 2018). 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 (Zhang et al., 2019).

Shipping emissions were also analysed for monthly and seasonal patterns. Seasonal amounts of shipping emissions for the pollutants analysed are shown in Table 4. According to 2015 data, the largest amounts of pollutants were emitted in summer and spring, accounting for approximately 26% of the annual total in both cases, being similar for the other seasons (23% and 25% for winter and autumn, respectively). Fig: 2 shows the monthly amounts of CO_2, NO_x and SO_x in tonne y^{-1} and ash, CO, EC, OC and sulphate in kg y^{-1} of shipping emissions in the study domain during 2015. 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). These slight variations seem to be related to the navigation conditions (better during the spring and summer), which consequently increases the number of ships that sails in this zone (during May, June, July and August). Fan et al. (2016) also reported slight seasonal variations similar to this study, although for East China Sea higher emissions were verified during spring. Jalkanen et al. (2009) reported higher shipping emissions during summer (highest emissions during July) for Baltic Sea in 2007 and a similar seasonal variation pattern, although the variation was higher (20% between the months with the highest and lowest NO_x emissions).

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3.2 Impact on Air Quality

Annual average concentrations

To understand shipping emissions impact on air quality over the Iberian Peninsula in 2015, EMEP model was configured considering and not considering shipping emissions. Fig: 3 shows the contribution of shipping emissions to the annual average of NO_2, SO_2, sulphate, O_3, $PM_{2.5}$ and PM_{10} surface concentrations in the Iberian Peninsula. Results show that when shipping emissions were considered, the concentrations of NO_2, SO_2, sulphate, $PM_{2.5}$ and PM_{10} increased, especially in the Strait of Gibraltar and close to the coastal areas (mainly in port areas) as well as along the west coast of the Iberian Peninsula, (along main shipping routes). O_3 concentrations also increased due to shipping emissions especially in the Mediterranean Sea close to the African coast. An opposite behaviour was verified with a decrease of concentrations around the major shipping routes in the west coast of the Iberian Peninsula, close to the southern coastal area of Spain and in some port areas as a result of NO_x titration caused by increased NO_x shipping emissions. Aksoyoglu et al. (2016) also reported an increase in the mean O_3 concentrations of 5–10 % in the Mediterranean Sea and a decrease of the levels around some major ship lanes (English Channel and North Sea). Moreover, Merico et al. (2016) that performed experimental measurements in a port-city in Italy found correspondence between NO peaks and O_3 titration. Thus shipping emissions have the potential to decrease O_3 concentrations close to the main ship lanes and ports and increase at larger distances from the emissions source, which seems to be a local scale effect. Annual mean concentrations when shipping emissions were included (considering all grid cells of the domain) of NO_2, SO_2, sulphate, O_3, $PM_{2.5}$ and PM_{10} were, respectively, 1.8 µg m^{-3}, 0.5 µg m^{-3}, 0.8 µg m^{-3} (mean increase of 67%), 80 µg m^{-3}, 8.2 µg m^{-3}, 22 µg m^{-3}.

Comparison with previous studies in the region

The highest differences in the annual mean concentrations of NO_2, SO_2, sulphate, O_3, $PM_{2.5}$ and PM_{10} w/ship case and wt/ship case were 31.7 µg m^{-3}, 16.1 µg m^{-3}, 3.4 µg m^{-3}, 13 µg m^{-3}, 4.8 µg m^{-3} and 6.9 µg m^{-3}, respectively. Monteiro et al. (2018) reported for Europe similar differences of PM_{10} (7 µg m^{-3}) and of O_3 (14 µg m^{-3}) and lower of NO_2 (18 µg m^{-3}) evidenced in the main shipping routes of Straits of La Mancha and Gibraltar. The higher NO_2 concentrations reported in this study compared with Monteiro et al. (2018) seem to be related to the shipping emissions inventory that was used. As was already mentioned the shipping emissions used by Monteiro et al. (2018) were extracted from TNO-MACC_III inventory which seems to underestimate NO_x emissions of this sector compared with the STEAM (more precise). Aksoyoglu et al. (2016) reported lower differences for $PM_{2.5}$ (3.5 µg m^{-3}) and O_3 (12 µg m^{-3}) for Europe. Fig: 4 shows the relative impact of shipping emissions on pollutant concentrations for the Iberian Peninsula. Locally, the effects were more evident in the sea areas along main shipping routes and especially in the Strait of Gibraltar and in the Mediterranean Sea, with contributions of more than 90% for NO_2 and SO_2, 80% for sulphate, 25-50% for $PM_{2.5}$ and 20-35% for PM_{10}. Regarding O_3, shipping emissions contributed to an increase of around 15% all over the Iberian Peninsula coastline and in the Mediterranean Sea close to the African Coastline. Nevertheless, shipping emissions also contributed to decrease O_3 concentrations around 15-40% in the Strait of Gibraltar and close to Valencia Port. It is also important to emphasise that along main shipping routes and close to the Iberian Peninsula port areas (except Valencia), O_3 concentrations considering and not considering shipping emissions remained the same. Aksoyoglu et al. (2016) found higher contributions (45%) for $PM_{2.5}$ and lower positive contributions (5-10%) for O_3 in the Mediterranean Sea. Sotiropoulou and Tagaris (2017) also reported contributions higher than 90% for NO_2 and SO_2 and 40% during winter and 50% during summer for $PM_{2.5}$ over the Mediterranean Sea. Viana et al. (2014) reviewed studies concerning the impact of shipping emissions on air quality in European coastal areas and reported lower contributions than those estimated in this study

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for the Strait of Gibraltar (2-4% for mean annual PM_{10} and 14% for mean annual $PM_{2.5}$) and Southern Spain close to Bay of Algeciras (3-7% for mean annual PM_{10} and 5-10% for mean annual $PM_{2.5}$). The differences between the contributions reported by Viana et al. (2014) seem to be related to the methodology used in the reviewed studies (source apportionment of PM_{10} and $PM_{2.5}$ by positive matrix factorization). Although the impact of shipping emissions on pollutants' concentrations has been most evident in sea areas, they also contributed to increasing inland concentrations. As shown in Fig: 4, shipping emissions generally contributed to about 50% of inland NO_2 concentrations near port areas of Portugal and Spain, reaching more than 75% in the province of Cadiz. Similar behaviour was observed for SO_2 concentrations, however, in this case, contributions of more than 75% were also noticed in the province of Malaga. As already mentioned, for O_3, contributions of around 5-15% were calculated for the entire Iberian Peninsula coastline, especially in the south coastal region. Regarding sulphate, contributions of around 60% were calculated for all the Iberian Peninsula south coastal region, with contributions of 20-40% when all the Iberian Peninsula was considered. For $PM_{2.5}$ and PM_{10}, the highest contributions (around 20-30%) were also verified in the Iberian Peninsula south coastal region. When all the Iberian Peninsula was considered, $PM_{2.5}$ and PM_{10} contributions were 10% and 15%, respectively. 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 underestimate PM_{10} and NO_2 concentrations (negative MBE), while the model used in the present study overestimate the concentrations (positive MBE).

Seasonal variation

The higher contributions of shipping emissions for pollutant concentrations in coastal regions (mainly to NO_2 and SO_2 concentrations) as well as in inland regions (sulphate, O_3, $PM_{2.5}$ and PM_{10} concentrations) indicates that ships are a non-negligible source. Regarding the seasonal concentration data, statistically significant variations were found for all pollutants across all seasons (*p*-values < 0.05). In fact, according to the model results, the higher contributions of shipping emissions to the concentrations levels were registered during spring and summer periods (warm season). This pattern seems to be related to the increase in ship traffic during summer due to better meteorological conditions that allow better navigation conditions, which increases the traffic and subsequently the emissions and atmospheric pollution. Moreover, during summer months, the number of passenger ships tends to increase (due to recreational travel), especially in the Mediterranean Sea, which led to an increase of shipping emissions and their contributions to the pollutant's concentration levels. Results were consistent with those achieved by Aksoyoglu et al. (2016), Chen et al. (2017), Sotiropoulou and Tagaris (2017) and Chen et al. (2018), which also reported largest contributions of shipping emissions on $PM_{2.5}$, O_3, NO_2 and SO_2 concentrations during summer.

Ship impact on exceedances of regulatory air quality limits

Fig: 5 shows NO_2, SO_2, O_3, $PM_{2.5}$ and PM_{10} exceedances to EU air quality standards and WHO air quality guidelines in the inland regions due to shipping, as well as the differences between SOMO35 levels (in ppb.days) considering and not

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considering shipping emissions. Results showed no exceedances to EU annual limit standard for SO_2, $PM_{2.5}$ and PM_{10}. Regarding NO_2, as the annual limit for the EU air quality standards and the WHO air quality guidelines are the same (40 µg m^{-3}), the analyses were joined. As can be seen from Fig: 5 a), exceedances due to shipping emissions were verified in Valencia area close of Valencia Port and in Barcelona area close of Port of Barcelona. When shipping emissions were considered $PM_{2.5}$ WHO air quality guideline (10 µg m^{-3}) exceedances increased 7%. As can be seen from Fig: 5 b), exceedances were verified in Portugal (close to the areas of Ports of Viana do Castelo, Leixões, Lisboa and Setúbal), across all Spanish coastline, in the north (in Pontevedra Province close to the area of Port of Vigo and in Asturias Province close to the areas of ports of Aviles and Gijon) and in the south (in the regions of Andalusia close to the areas of Algeciras, Malaga and Adra Ports, in Valencia close to area of Port of Valencia and in the Catalonia close to the area of Port of Barcelona) where the contribution of shipping emissions to the increase of concentrations was even more pronounced. It should be noted that shipping emissions were still responsible for exceedances in areas far from the coastline, as was verified in Viana do Castelo and more pronounced in the region of Andalusia. These results confirm that shipping emissions can contribute negatively to air quality, both in coastal and in inland areas. PM_{10} WHO air quality guideline of 20 µg m^{-3} was exceeded 8% more when shipping emissions were considered. As can be seen from Fig: 5 c), exceedances were verified mainly across the southern Spanish coastline, in the regions of Andalusia and Catalonia. The contribution of shipping emissions to the increment of number of exceedances (in terms of concentrations Δ µgm^{-3}) of NO_2, $PM_{2.5}$ and PM_{10} was also determined. This information can be found in Fig: A2 a), b) and c), respectively. Regarding WHO air quality guideline for SO_2, as the value refers to the average daily concentrations, the results are presented as the number of days per year that the threshold value was exceeded in a given grid cell when shipping emissions were considered but were not exceeded without shipping emissions. Fig: 5 d), shows that exceedances were verified in Setúbal District (Portugal) close to the area of Port of Sines, across all Spanish coastline, in the north (in Coruña Province close to the area of Port of Coruña, in Asturias Province close to the area of ports of Aviles and Gijon and close to the area of Port of Bilbao) and in the south (in Huelva close to the area of Port of Huelva, in Cadiz Province close to the Strait of Gibraltar, in Valencia close to the area of Port of Valencia, in Castellón close to the area of Port of El Grao, in Tarragona close to the area of Port of Tarragona and in Barcelona close to the area of Port of Barcelona). In the Strait of Gibraltar, it was calculated the highest number of days per year where WHO reference value for SO_2 was exceeded (maximum increment of 96 days). The spatial distribution of the number of days per year in which the WHO reference value for SO_2 was exceeded can be found in Fig: A2 d). According to the above results, mitigations measures should be studied and implemented to reduce shipping emissions mainly close to port areas, in the south of the Iberia Peninsula close to the Strait of Gibraltar and in the Mediterranean Sea. Implementing an ECA in the Mediterranean Sea can contribute to reduce shipping emissions and help these regions to attain WHO and EU standards. As SOMO35 is an indicator of health impact assessment recommended by WHO, differences between the levels considering and not considering shipping emissions were calculated to evaluate the contribution of these emissions for the O_3 inland concentrations. As it can be seen from Fig: 5 e), SOMO35 levels were negative close to the Portuguese the areas of ports of Lisboa and Setúbal and close to Spanish the areas of ports of Algeciras (Strait of Gibraltar), Valencia and Barcelona. The major contributions were calculated for the southwest coastline of the Iberian Peninsula, with

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levels from 500-1000 ppb.days up to 200 km from the coastline (over all south region of Portugal), which might be explained by the highest solar radiation intensity that is felt in the southern regions of the Iberian Peninsula.

3.3 Uncertainties and Limitations

Given the complexity of any chemical transport model, it is difficult to specify the source of uncertainties, these are inherent to the uncertainties of the meteorological data, emission inventory and the imperfections of chemical mechanism and physical process on the modelling system. Nevertheless, it is known that the reliability of the emissions inventory is a major cause of uncertainty. Efforts to reduce uncertainties were made by using shipping emissions input data as accurate as possible, estimated by an improved version of STEAM model (STEAM3), that has the highest spatially detailed shipping emissions inventory and have been recognized as one of the best to estimate emissions from maritime traffic (Nunes et al., 2017b; Russo et al., 2018). Keeping the uncertainties of the atmospheric dispersion simulations in mind, efforts were made to run the EMEP-MSC/W model as accurate and detailed as possible (horizontal resolution of 0.1°x0.1°, 34 vertical levels and data output time steps of 1 h). 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 NO_2, SO_2 and $PM_{2.5}$ resulting of shipping emissions, although it seems to be underestimating $PM_{2.5}$ concentrations and overestimating O_3 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. The EMEP-MSC/W model considers the O_3 loss by NO_x titration, the sunlight effects and NO_x to VOC ratio that promotes O_3 production, which is an approximation allowing to minimize the effects of the non-linear O_3 chemistry. Moreover, estimations were performed using meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) for 2015.

4 Conclusions

In this study, Ship Traffic Emission Assessment Model (STEAM3) was used to estimate shipping emissions in the Iberian Peninsula Region in 2015. According to the results, total estimated emissions for CO, CO_2, SO_x, NO_x and particulate matter (subdivided in elementary carbon (EC), organic carbon (OC), sulphates and ash) were 49, 30000, 360, 710, 4.5, 11, 32 and 3.3 ktonnes y^{-1}, respectively. The highest emissions were estimated along the west coast of the Iberian Peninsula, in the Strait of Gibraltar and in the Mediterranean Sea. The largest amount of emissions for all pollutants were emitted during summer and spring (reaching the maximum during July) which seemed to be related to the navigation conditions. The estimated shipping emissions were equivalent to 45% and 62% of NO_x and SO_x of the total land-based emissions, respectively, which shows that shipping emissions cannot be neglected. Running the EMEP/MSC-W model it was possible to observe that the effects of shipping emissions on air quality were more evident in the sea areas along the main shipping routes and especially in the Strait of Gibraltar and in Mediterranean Sea. Although the contribution of shipping emissions to pollutants concentrations has been more evident in sea areas, they also contributed to increasing the inland concentrations. It was observed that shipping emissions

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increased SO_2 and NO_2 concentrations around 50% near port areas of Portugal and Spain, reaching more than 75% in the provinces of Cadiz and Malaga, O_3 concentrations around 5-15% for all the Iberian Peninsula coastline, especially in the south coastal region and sulphate, and particulate matter ($PM_{2.5}$ and PM_{10}) concentrations around 60% and 20-30%, respectively all over the Iberian Peninsula south coastal region. NO_2, exceedances due to ship emissions were detected in Valencia and Barcelona. WHO air quality guideline for $PM_{2.5}$ and PM_{10} were exceeded, respectively, 7% and 8% more when shipping emissions were considered. In the regions close to the Strait of Gibraltar it were observed the highest exceedances of WHO air quality guideline for SO_2 (maximum increment of 96 days). The major contributions of shipping emissions to inland SOMO35 levels were for the southwest coastline of the Iberian Peninsula, with levels of 500-1000 ppb.days up to 200 km from the coastline (overall south region of Portugal). These results confirm that shipping emissions can contribute negatively to air quality, both in coastal and in inland areas and mitigations measures should be studied and implemented to reduce shipping emissions mainly close the port areas and in the south of the Iberia Peninsula (close to the Strait of Gibraltar and in the Mediterranean Sea). In the future, it is important to study the impacts of shipping emissions on health, which are still underestimated and rarely studied.

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Code availability

The EMEP model is available as Open Source (see https://github.com/metno/emep-ctm code version rv4.17 (201802) (EMEP MSC-W, 2018, https://doi.org/10.5281/zenodo.3355023). STEAM model is intellectual property of the Finnish Meteorological Institute and is not publicly available.

Author contributions

RAON performed EMEP MSC-W simulations and the analysis, did the interpretation of the results and wrote the manuscript. JPJ and HH provided the ship emission data. MCMA, FGM, JMG and JPJ reviewed the paper and helped in the interpretation of the results. FCC, VDG, and JMG gave support in the interpretation of the results for Spain and reviewed the paper. SIVS designed the study and assisted in modelling scenarios and in writing the paper.

Competing interests

The authors declare that they have no conflict of interest.

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Table 1. Model quality indicators for the present study estimations 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

Table 2. Annual mean amounts of emitted air pollutants from shipping and from land-based anthropogenic sources during 2015 (in tonne y^{-1})

Deleted: 1

Pollutant	Shipping	Land-based emissions [a]	Road transport emissions
Ash	3.3E+03	-	-
EC	4.5E+03	-	-
OC	1.1E+04	-	-
NO_x	7.1E+05	1.6E+06	6.6E+05
SO_x	3.6E+05	5.8E+05	7.1E+03
Sulphate	3.2E+04	-	-
CO_2	3.0E+07	-	-
CO	4.9E+04	3.6E+06	5.7E+05
Total	3.1E+07	-	-

[a] Emissions from 11 SNAP sectors, namely, public electricity and heat production, industry, other stationary combustion sources, fugitive emissions, solvents, road transport, aviation, off-road sources, waste, agriculture livestock, agriculture other sources and other sources.

Table 3. Annual average and highest intensities of NO_x and SO_x (in tonnes/yr/km^2) reported from researches in Asian Region.

Study	**Port/sea area**	**NO_x**		**SO_x**	
		Annual average	**Highest value**	**Annual average**	**Highest value**
Chen et al. (2016a)	Tianjin Port	5.06	1.51E+03	7.14	1.79E+03
Chen et al. (2017)	Qingdao Port	1.83	-	1.42	-
Fan et al. (2016)	East China Sea	1.0	1.0E+04	1.90	1.30E+03
Ng et al. (2013)	Hong Kong	-	1.1E+02		2.0E+02

Table 4. Seasonal amounts of emitted air pollutants from shipping in the Iberian Peninsula in 2015 (in tonne y^{-1})

Pollutant	Spring	Summer	Autumn	Winter	Total
Ash	0.85	0.87	0.83	0.77	3.3
EC	1.2	1.2	1.1	1.0	4.5
OC	2.9	3.0	2.8	2.6	11
NO_x	1.8E+02	1.9E+02	1.8E+02	1.6E+02	7.1E+02
SOx	92	94	91	85	36
Sulphate	8.3	8.4	8.1	7.6	32
CO_2	7.8E+03	8.0E+03	7.6E+03	7.0E+03	3.0E+04
CO	13	13	12	12	49
Total	8.3E+03	8.3E+03	7.9E+03	7.2E+03	3.1E+04

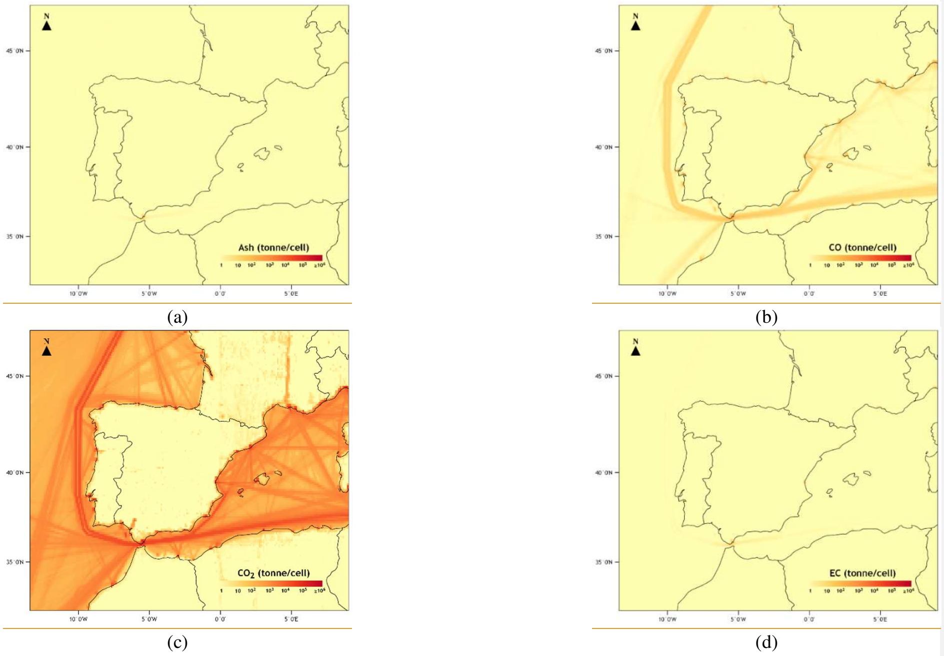
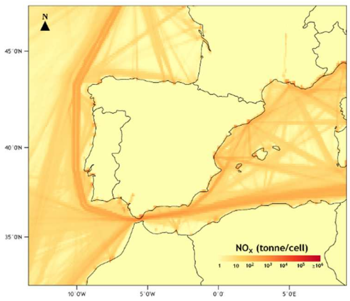
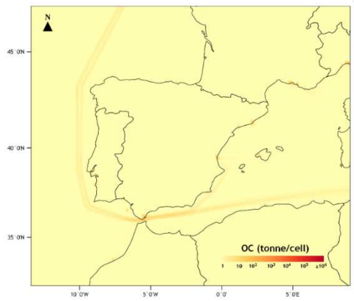


Figure 1: Shipping emissions of a) ash; b) CO; c) CO_2; d) EC; e) NO_x; f) OC; g) sulphate and h) SO_x in the study domain for 2015



(e)



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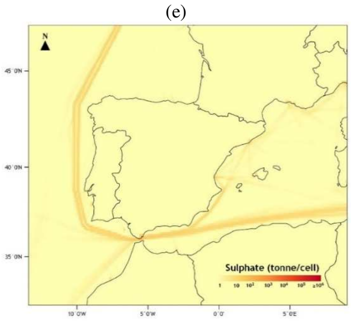

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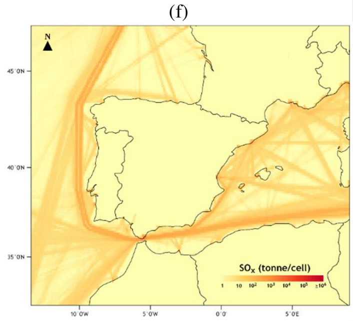

(h)

Figure 1 (continued): Shipping emissions of a) ash; b) CO; c) CO_2; d) EC; e) NO_x; f) OC; g) sulphate and h) SO_x in the study domain for 2015

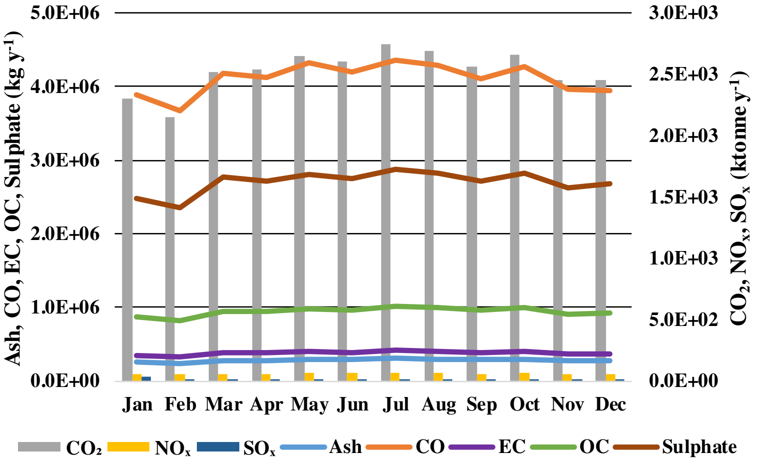

Figure 2: Monthly amounts of CO_2, NO_x and SO_x in ktonne y^{-1} (bars-right axis) and ash, CO, EC, OC and sulphate in kg y^{-1} (lines-left axis) shipping emissions in the study domain during 2015

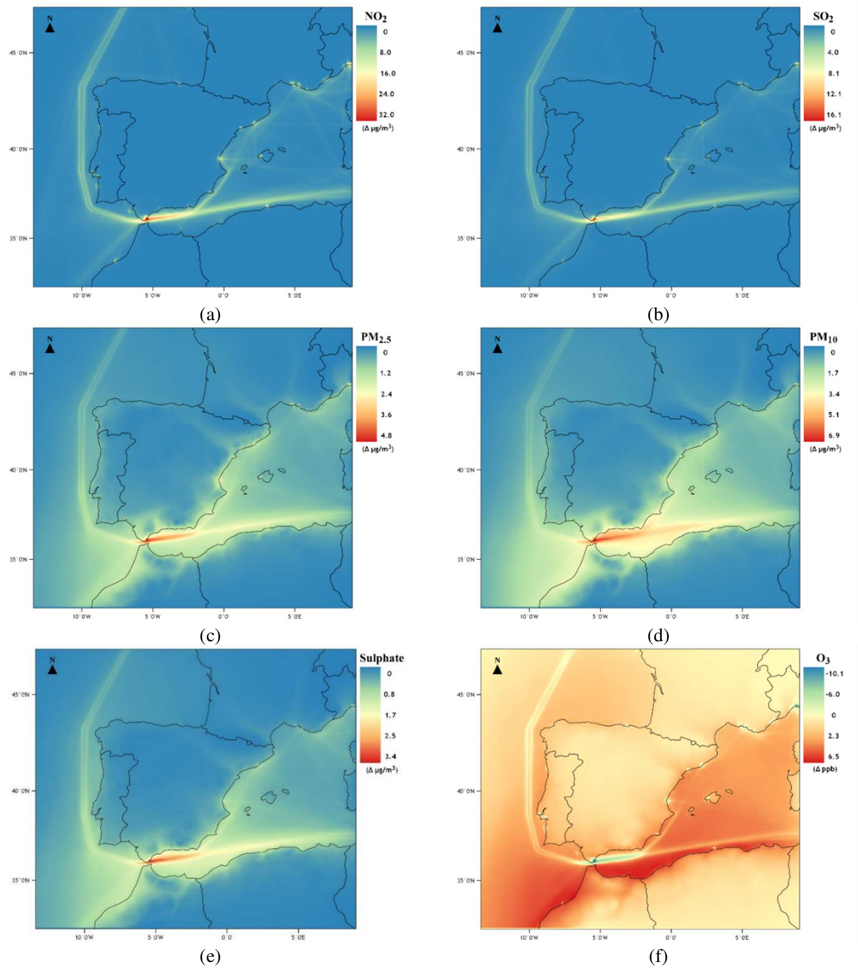

Figure 3: Contribution of shipping emissions to annual mean surface concentrations of a) NO_2; b) SO_2; c) $PM_{2.5}$; d) PM_{10}; e) sulphate; f) O_3 in the study domain in 2015 (Δ=S-SCN - B-SCN)

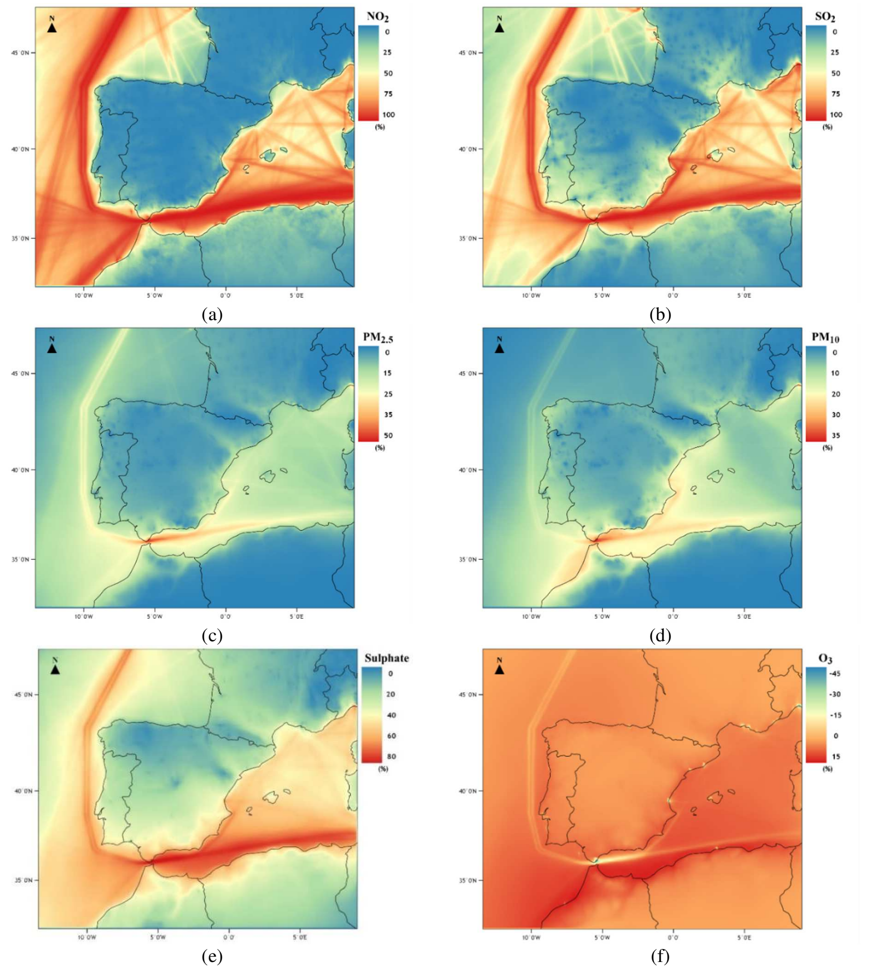

Figure 4. Contribution of shipping emissions (%) to annual mean concentrations of a) NO_2; b) SO_2; c) $PM_{2.5}$; d) PM_{10}; e) sulphate and f) O_3 for 2015

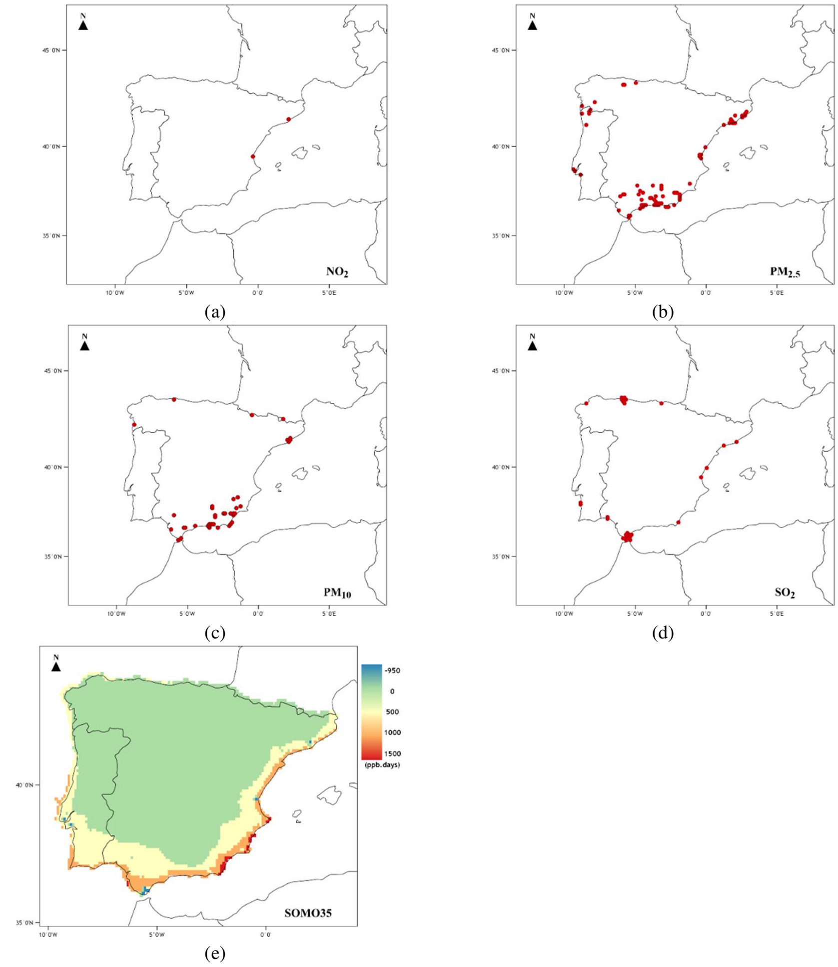

Figure 5: Spatial distribution of the inland exceedances for a) NO_2 to EU air quality standards and WHO air quality guidelines (same value); b) $PM_{2.5}$; c) PM_{10} and d) SO_2 to WHO air quality guidelines and e) SOMO35 levels due to shipping emissions contributions.

Appendix A

This appendix contains the spatial distribution of world shipping traffic density and a zoom of the study area for 2015, as well as the spatial distribution of the contribution of shipping emissions in terms of concentration increment to the inland exceedances.

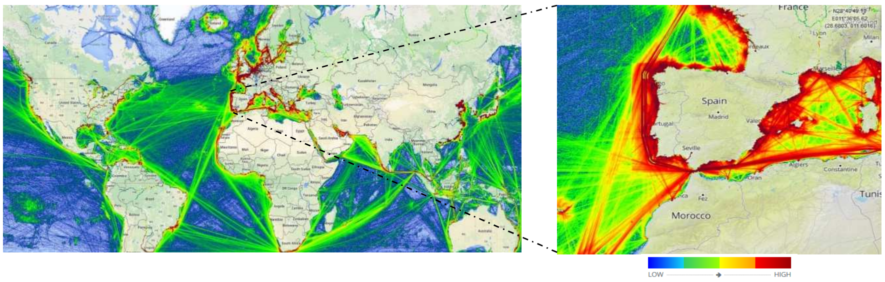

Figure A1. World shipping traffic density map and a zoom of the study area for 2015 (source: Marine Traffic, 2016).

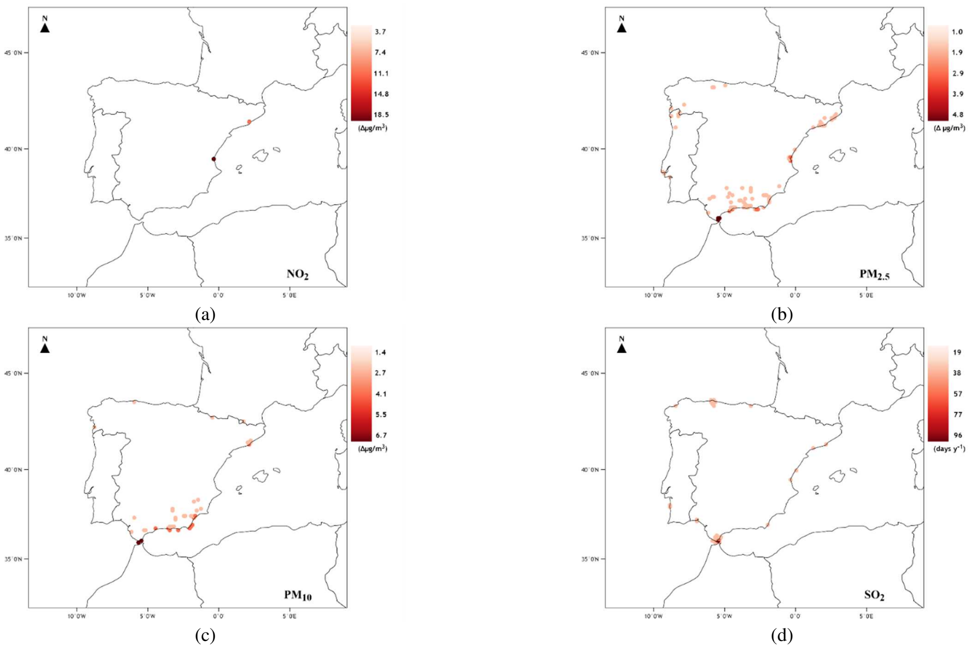

Figure A2. –Spatial distribution of the contribution of shipping emissions (concentration increment) to the inland exceedances for a) NO_2; b) $PM_{2.5}$ and c) PM_{10} and number of days per year that the threshold value was exceeded in a given grid cell for d) SO_2.