Dear Editor and Referees,

We are pleased to submit our responses to all the comments and revision for manuscript acp-2018-1163. We appreciate all the comments and suggestions that are especially helpful. All the referees' comments have been addressed carefully.

Best regards with respect,

Yan Zhang, representing all co-authors

Reviewers' comments are in blue.

Authors' responses are in black.

Revisions in manuscript are in *italic*, underlined.

Response to Referee's Comments #1

General comments:

1. The manuscript of Feng et al, "The influence of spatiality on shipping emissions, air quality and potential human exposure in Yangtze River Delta/Shanghai, China", is well written and provides some additional information on the spatial distribution of ship emissions of the inland waterway traffic. This manuscript feels like an attempt to achieve something greater in the future, because it introduces the methodology necessary for ship emission inventory work, atmospheric modeling and health effect evaluation without getting there in the end. The title wisely stops at human exposure, because this is what the paper delivers, but I wonder why the authors stopped there and did not take the final step from exposure to health effects.

Response:

We thank the reviewer for this comment. As the reviewer has noted, the focus of this paper was on the results of our approach to estimating the impact of shipping and related activities on $PM_{2.5}$ concentrations and where those concentrations differed when examined in light of where the population lives. We do intend to prepare a manuscript that examines the health impacts of these exposures in detail, and have added to the text that "this work only extends from emissions to air quality and population exposures. The health impacts of shipping-related air pollution in Shanghai and the YRD region will be explored in future work."

Revisions in the manuscript:

1. Page 20, line 8-10: "Finally, this work only extends from emissions to air quality and population exposures. The health impacts of shipping-related air pollution in Shanghai and the YRD region will be explored in future work."

2. The novelty aspect of this work could be improved; emission inventory work cites existing work and this paper does not bring much new to this topic. The atmospheric modeling was done with an existing code and no advances were made to improve the existing tools. From methodological point of view, this paper applies existing tools to

a known environmental problem which means that the novelty must come from that contribution. There are two contributions which are brought to light in this paper. First is the contribution of inland waterway traffic to ship emissions and the second is the geographical reach of ship emissions when ship to shore distance is varied. The latter contribution hints to a design of new potential regulation which would not necessarily cover all of the 200 nautical mile distance from shore, but this motivation is currently only indirectly stated, if at all.

Response:

Thank you for the suggestions to more clearly identify the novelty of our work, especially regarding the contribution of inland waterway traffic to ship emissions and the spatial distribution of ship emissions, ambient pollutant concentrations, and human exposures. We have expanded on the description of the gap in the literature that this paper addresses and the policy implications of the research results throughout the manuscript. Also, it is also novel that both of the regional and port scale influences of shipping emission on air quality have been considered in this manuscript.

Revisions in the manuscript:

1. Page 2, line 6-8: "in particular, in the YRD region, expanding the boundary of 12 NM in China's current DECA policy to around 100 NM would include most of the shipping emissions affecting air pollutant exposures, and stricter fuel standards could be considered for the ships on inland rivers and other waterways close to residential regions."

2. Page 5, lines 6-15: "In China, a few studies reported the contribution to air pollution from shipping in different offshore coastal areas or different-type ship-related sources. For example, Mao et al. (2017) estimated primary emissions from OGVs at different boundaries in the PRD region, and concluded that further expansion of emission control area to 100 NM would provide even greater benefits. However, the impacts of shipping emissions at varying distances from shore on air quality and potential human exposure, which are important when considering ECA policy, have not been rigorously studied. Mao and Rutherford (2018) studied NO_x emissions from three categories of merchant vessels—OGVs, coastal vessels (CVs) *and river vessels (RVs) in China's coastal region. But less attention was paid to the impacts of inland waterway traffic and port-related sources like container-cargo trucks and terminal port equipment on air quality and potential human exposure."*

3. Page 5, line 30; page 6, line 1-3: "The results of this study could be informative to the consideration of the distance of regulated emissions in the design of future emissions control areas for shipping in YRD, or regulations on the sulfur content of fuels for different-type ship-related sources in Shanghai."

4. Page 16, line 3-8: "The results of these YRD analyses suggest that although ambient ship-related SO_2 concentrations were mainly affected by shipping inland or within 12 NM, expanding China's current DECA to around 100 NM or more would reduce the majority of the impacts of shipping on regional $PM_{2.5}$ pollution. It also implies that the future ECA policy should consider multiple air pollutants including the primary and secondary pollutants synchronically."

5. Page 19, line 10-12: "The results of the analyses of different-type shipping-related sources indicated that ship-related sources close to densely-populated areas contribute substantially to population exposures to air pollution."

6. Page 20, line 28-29; page 21, line 1-4: "For example, policymakers could consider whether to expand China's current DECA boundary of 12 NM to around 100 NM or more to reduce the majority of shipping impacts on air pollution concentrations and exposure. It will be helpful to improve the local air quality and reduce human exposures in densely populated areas by developing more stringent regulations on the fuel quality for ships entering inland rivers or other waterways close to residential regions."

3. In some parts of the manuscript, authors state that they have used data from specific months whereas on other parts data for a full year seems to be used. It was challenging to understand which parts of the work were done with a full year's dataset and which with less data.

Response:

Thank you for pointing out this problem. We have revised the relevant parts of the manuscript to clarify the temporal scope of data used in our work. It is due to the limitation of getting the national-scale AIS data for the whole year from the marine-time department, only data in some representative month like January and June are available for our study. Therefore, we used the average values of these two months to estimate annual shipping emissions in whole China. But we have full-year AIS data in Yangtze River Delta (YRD), and the estimates of annual shipping emissions in YRD scale and Shanghai city scale in the manuscript were based on the full-year data. To identify the impact of shipping on ambient air quality and population exposure, January and June were selected as representative months to conduct sensitivity experiments, and monthly shipping emissions for January and June were used in the air quality model. We have clarified the time scale of data we used in the revised manuscript.

Revisions in the manuscript:

1. Page 5, line 18-28: "We modeled shipping emissions in different offshore areas in the YRD region and emissions from different-type ship-related sources in Shanghai city for each month of the year. To identify which offshore areas in the YRD region and which ship-related sources in Shanghai contributed the most ambient air pollution, and human population exposure, we modeled the impacts of shipping emissions in different offshore areas (within 12 NM including inland waters, 12-24 NM, 24-48 NM, 48-96 NM, and 96-200 NM) in the YRD region as well as coastal ships, inland-water ships, and container-cargo trucks and port terminal equipment in and near the port areas under the jurisdiction of Shanghai MSA in two representative months (January and June)."

2. Page 7, line 9-11: "*Due to limitation of the data source, the national-scale AIS data in this study only covered the representative months of January and June 2015, while the YRD-scale AIS data covered 2015 full year.*"

3. Page 7, line 12-16: "Emissions from ships entering the geographic domains for YRD or Shanghai were calculated using the AIS-based model developed by Fan et al. (Fan et al., 2016), and monthly shipping emissions for January and June were used in the air quality model to capture the seasonal variation to expect more accurately than *annual shipping emissions with no monthly variations.*"

Detailed comments:

4. Page 1, Introduction, lines11-15. Authors discuss the health effect evaluation of ship emissions and quote Sofiev et al (2018). I wonder, what is the motivation of not citing the numbers of Sofiev et al, which reports the latest global health effect numbers, but authors choose to refer to 50 000 to 90 000 premature mortality cases instead? The values given in Sofiev et al (2018) are much higher than this.

Response:

Thank you for the question. Previously, we more focused on the impact of shipping in past years, so that the numbers we referred to are the estimates of past years (2010 and 2012). Based on your reminding, we think it is better to also cite the value in Sofiev et al (2018), which reports a 2020 projection of shipping's impact, so as to give a more comprehensive review of the health effect evaluation of ship emissions.

Revisions in the manuscript:

1. Page 2, line 19-24: "Globally, about 50,000 to 90,000 cardiopulmonary diseases and lung cancer deaths were attributable to exposure to particulate matter emitted from shipping in 2010 and 2012, respectively (Corbett et al., 2007;Partanen et al., 2013;Winebrake et al., 2009), and 403,300 premature mortalities per year due to shipping are predicted in 2020 under business-as-usual (BAU) assumptions (Sofiev et al., 2018)."

5. Page 4, lines 24-29. Authors have chosen to report the case before the DECA implementation. I was wondering about the motivation of this decision, because it seems that the modeling work could have been easily applied also the DECA case and would have allowed the identification of the impacts of this policy change thus significantly improving the novelty aspect of this work.

Response:

Thank you for the question. This study aimed to evaluate the impact of shipping emissions on air quality prior to implementation of the DECA policy as the baseline.

Taking 2015 as the baseline year can reflect the situation for recent years. Also, this research aimed to provide basic scientific evidence to inform policies for controlling future shipping emissions. The first-phase DECA policy during 2016-2018 only applied to ships during berthing at port. Evaluation of potential future DECA policies will be done in ongoing work.

6. Page 5, lines 15-22: Authors report the specifics of chemical transport model domains, but say very little of the emissions. There is a separate section for ship emissions, but I cannot see whether daily, monthly or annual emissions with or without the dynamic features of ship emissions were used or not. The activity data allows this, but have the authors considered these variations in to consecutive steps, too?

Response:

Thank you for the question. The ship emissions were actually calculated at 5-minute intervals based on AIS data. Then we used the monthly dynamic ship emissions as the input to the air quality modelling since our air quality analysis has been based on the monthly time scale. Also the monthly mean simulation results were evaluated in this study to show they can match with the observations well. We've clarified the use of monthly shipping emission in the air quality model in the revised manuscript.

Revisions in the manuscript:

1. Page 7, line 12-16: "Emissions from ships entering the geographic domains for YRD or Shanghai were calculated using the AIS-based model developed by Fan et al. (Fan et al., 2016), and monthly shipping emissions for January and June were used in the air quality model to capture the seasonal variation to expect more accurately than annual shipping emissions with no monthly variations."

7. Page 5, lines 23-27. "Highest shipping impacts were expected in June because shipping activity and emissions are higher in summer than at other times of the year". There are references to Fan et al (2016) and Jalkanen (2009) in this sentence. Actually

Fan et al state "No significant differences in the total emissions quantities were observed among summer, autumn and winter", which seems to contradict what the authors say.

Response:

Thank you for the question. In this study, shipping emissions in summer were slightly higher than the other seasons (The ship emission in summer accounted for more than 28% of the annual shipping emissions, a little higher than other season). In general, the variation in total emissions among different seasons was small, which is consistent with other studies (Corbett et al., 1999; Fan et al., 2016). Therefore, meteorological differences were the dominant factor affecting the seasonal differences of ship-related impacts on air quality. Therefore, we've modified this sentence into "higher shipping impacts were expected in June because prevailing winds from the summer monsoon are directed from the ocean to the shore, along with higher ship emissions in summer".

Revisions in the manuscript:

1. Page 6, line 28-29: "Two representative months in the year 2015, January and June, were selected to compare the seasonal effects. Higher shipping impacts were expected in June because prevailing winds from the summer monsoon are directed from the ocean to the shore."

8. Page 6, lines 1-2. I would like to see some discussion on the limitations of AIS in this. It is not used by all ships listed by the authors. The way the text is written now implies that all the ship classes listed here is covered by AIS, which is not necessarily the case since inland traffic may be incompletely represented.

Response:

Thank you for the suggestion. The way the text was written was not fully appropriate and we've modified the sentence into "AIS data includes international ships, coastal ships, and inland-water ships, but some river ships could be not covered by AIS data". In addition, we have added section 3.4 Limitations to address the limitations of AIS data as a part of it.

Revisions in the manuscript:

1. *Page 7, line 11-12: "AIS data includes international ships, coastal ships, and inland-water ships, but some river ships could be not covered by AIS data."*

2. *Page 19, line 13-29; page 20, line 1-7:*

*"**3.4 Limitations and uncertainties***

Limitations in the study were mainly related to some missing information in database and assumptions during estimation of shipping emissions. When estimating shipping emission inventory, underestimations of actual emissions may be introduced by missing information. For example, AIS data has a high coverage of coastal vessels, but many inland vessels are not equipped with AIS. Emissions from those inland vessels without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA, and that could introduce some uncertainties for inland river vessels. Also, emissions from fishing boats were probably underestimated because AIS devices on some fishing boats may not be in use. Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions.

In addition, we did not consider the external effects of water flow, wind, and waves when calculating engine power for ships going over the region. This would introduce some uncertainties (Aulinger et al., 2016). According to the previous studies in other areas, these factors may increase fuel consumption of individual vessels by as much as 10% to 20%, while the effects of waves on emissions estimations over extensive geographical regions are negligible (Jalkanen et al., 2009; Jalkanen et al., 2012). The downstream of the Yangtze River is located in the geographically plateau region, and the river flow is below 0.5 m/s (Song and Tian, 1997; Xue et al., 2004). For Shanghai, located at the end of mouth of the Yangtze River to the East China Sea with a flat terrain, the river flow is very slow. Given that ships traveling the Yangtze River near Shanghai have speeds over ground (SOG) of about 5-10 knots (3-5 m/s), the relative ratios of water flow to the SOG is within 20%.

In our future work, we will fill the gap in the basic ship data and consider the external effects when building the shipping emission inventory."

9. Page 6, lines 4-6. Does the material obtained from MSA include boats? I would imagine that boats outnumber ships by at least an order of magnitude. Was any consideration given to boat contributions to emissions? Boats may not be the biggest source of CO2, NOx or SOx, but they are a significant source of VOCs and CO.

Response:

Thank you for the question. The material, 2015 vessel call data, obtained from MSA includes information on some registered inland boats. However, emissions from fishing boats were probably underestimated, since some AIS devices on fishing boats may not be in use. Discussion on underestimation of emissions from fishing boats has been added to the new section on limitations.

Revisions in the manuscript:

1. *Page 19, line 15-24:* "*When estimating shipping emission inventory, underestimations of actual emissions may be introduced by missing information. For example, AIS data has a high coverage of coastal vessels, but many inland vessels are not equipped with AIS. Emissions from those inland vessels without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA, and that could introduce some uncertainties for inland river vessels. Also, emissions from fishing boats were probably underestimated because AIS devices on some fishing boats may not be in use. Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions.*"

10. Page 6, lines 7-12. Use of speed entries of AIS. How did you count for the water flow? You have concentrated the study on an area which is along a large river, which

means that there is a significant water flow. When a case like this occurs, speed over water is not the same as the speed over ground indicated by the AIS. If power predictions are based on speed over ground, then power prediction will fail. Have the authors considered this aspect?

Response:

Thank you for pointing out the potential importance of water flow. The Yangtze River is indeed a large river with the length of 6300 km, divided into the upstream, middle stream and the downstream. The average river flow rate of the upstream and middle stream is in the range of 0.5-4.5 m/s due to the great height difference (Li, 2016; Xue et al., 2004). But for the downstream region, the geographically plateau region, the river flow is below 0.5 m/s (Song and Tian, 1997; Xue et al., 2004). For Shanghai, located at the end of the Yangtze River with a flat terrain, the river flow is very slow. Given that ships traveling the Yangtze River near Shanghai have speeds over ground (SOG) of about 5-10 knots (3-5 m/s), the relative ratios of water flow to the SOG is within 20%. We did not consider the influence of the water flow when calculating engine power for ships in this area for now. But your suggestions will be very useful for our further work extended for larger domain covering the middle and upstream Yangtze River. Also, we have added some discussion of water flow to the new section on limitations in Section 3.4.

Revisions in the manuscript:

1. Page 19, line 27-29; page 20, line 1-5: "*In addition, we did not consider the external effects of water flow, wind, and waves when calculating engine power for ships going over the region. This would introduce some uncertainties (Aulinger et al., 2016). According to the previous studies in other areas, these factors may increase fuel consumption of individual vessels by as much as 10% to 20%, while the effects of waves on emissions estimations over extensive geographical regions are negligible (Jalkanen et al., 2009; Jalkanen et al., 2012). The downstream of the Yangtze River is located in the geographically plateau region, and the river flow is below 0.5 m/s (Song and Tian, 1997; Xue et al., 2004). For Shanghai, located at the end of mouth of the Yangtze River to the East China Sea with a flat terrain, the river flow is very slow.*

Given that ships traveling the Yangtze River near Shanghai have speeds over ground (SOG) of about 5-10 knots (3-5 m/s), the relative ratios of water flow to the SOG is within 20%. In our future work, we will fill the gap in the basic ship data and consider the external effects when building the shipping emission inventory."

11. Page 6, lines 13-14. This is a rather drastic assumption. Have you thought about linking the fuel type or S content to engine specifications? There are technical reasons why some engines cannot use certain types of fuels, but have authors chosen to neglect these limitations completely?

Response:

Thank you for the question. Sulfur content is related to fuel type, and some engines can only use certain fuel types. In this study, the fuel type and sulfur content were linked to engine specifications in the model to estimate shipping emission (Fan et al., 2016). We have clarified the link among sulfur content, fuel type and engine type in the supporting information.

Revisions in the manuscript:

1. Page 7, line 24-26: "Assumptions regarding the fuel types, sulfur contents and engine types, and sources of emission factors, low load adjustment multipliers, and control factors are provided in section S.2 of the supporting information."

2. Supporting information, Page 3, line11-25: "The two most common fuel oils used in ships are residual oil (RO) and marine distillates (MD). In general, RO is used in the main engine, and the fuel sulfur content is approximately 2.7%, MD is used in the auxiliary engine, and the sulfur content is approximately 0.5%. On the basis of data on ships passing by the Port of Shanghai provided by the largest Chinese heavy fuel oil (HFO) supplier, China Marine Bunker (CMB), the sulfur content of the fuel used by the main engines in domestic vessels ranges from 0.2% to 2.0%, and the sulfur content of the fuel used by the main engines in ocean-going vessels ranges from 1.9% to 3.5%. In this study, we adjusted the sulfur content of the fuel used by the main engines in domestic vessels to 1.5% and that of ocean-going vessels to 2.7%. The amount of SO_2 *emitted is directly affected by the sulfur content of the fuel; therefore,* *when main engine emissions were estimated by the model, the emissions of domestic vessels were amended correspondingly. The main engine category was sorted into slow speed diesel (SSD), medium speed diesel (MSD), and high speed diesel (HSD) based on the engine revolutions per minute (RPM), and the largest auxiliary engine category was MSD. The type of engine was judged first according to the RPM of the main engine in Lloyd's database. The emission factors of the different types of engines differ considerably."*

12. Page 7, lines 3-6. "Shopping" of emission data piece by piece from various data providers may lead to unexpected side effects, which can arise from the fundamental assumptions used in emission inventory construction work. The CO and VOC emissions both result from incomplete combustion of fuel and there is a high probability that these two are linked. Did the authors check what the CO/VOC share in IIASA inventory was and how different the CO/VOC share was in the combined inventory?

Response:

Thank you for the question. We've checked the CO/VOC share in both IIASA inventory and the combined inventory. For the national scale land-based emission inventory, the emissions of CO and VOC in IIASA inventory are 1.84×10^5 kt/yr and 2.39×10^4 kt/yr, respectively, and the CO/VOC share is 7.7. The emissions of CO and VOC in the combined inventory are 1.79×10^5 kt/yr and 2.37×10^4 kt/yr, respectively, and the CO/VOC share is 7.5. Therefore, the CO/VOC share in the combined inventory is very close to the one in IIASA inventory. Besides, the CO emission from IIASA inventory was only used for national-scale nested domain in the air quality modelling (domain 1: the whole China, and domain 2: East China). For local-scale domain in the air quality modelling (domain 3: the YRD, and domain 4: Shanghai city), Shanghai Academy of Environmental Science (SAES) provided the complete land-based emission database.

Revisions in the manuscript:

1. Page 8, line 21-26: "In case of the large uncertainty leaded by merging data from two datasets, the ratio of CO to VOC was checked in this study. CO and VOC emissions both result from incomplete combustion of fuel and are likely to be related (von Schneidemesser et al., 2010; Wang et al., 2014). The ratio of CO to VOC was 7.7 in the IIASA inventory and 7.5 in the final combined inventory. Thus, the CO/VOC shares in these two inventories were very close and the use of the final combined inventory is acceptable."

13. Page 7, lines 16-21. What was the temporal resolution of the ship emission inventories used in this work?

Response:

Thank you for the question. The monthly ship emission inventories were used in this work. We've clarified this in the revised manuscript.

Revisions in the manuscript:

1. Page 7, line 12-16: "Emissions from ships entering the geographic domains for YRD or Shanghai were calculated using the AIS-based model developed by Fan et al. (Fan et al., 2016), and monthly shipping emissions for January and June were used in the air quality model to capture the seasonal variation to expect more accurately than annual shipping emissions with no monthly variations."

14. Page 8, lines 15-17, the last sentence. There is no uncertainty involved in atmospheric measurements? Really? These can be tens of percent, easily. Cross comparisons of AQ measurement results between instruments can deviate significantly, depending on the equipment used.

Response:

Thank you for the question. Indeed, uncertainties could exist in the measurement data, and we have revised this sentence.

Revisions in the manuscript:

1. Page 10, line 10-15: "The deviations between the simulation results and the monitoring data were mainly due to the uncertainties of emission inventories and *some deficiencies of meteorological and air quality models.* *<u>However, there were also uncertainties associated with the measurements themselves and the comparison of grid-based predictions to measurements at point locations.</u>"*

15. Page 8, lines 25-29. I agree that population weighted approach has some merit, but that still is an incomplete representation of human activity. The approach used here assumes that people spend all their time at home and do not consider realistic behavior of people. There are some studies that take this into account (see for example Soares et al, GMD, 2014).

Response:

Thank you for the question. The reviewer is correct that population-weighted exposures are an approximation of exposure given the more complex reality of where people spend their time.

Soares et al. represent a class of methods that have been in the literature for many years but that have rarely been applied on a large population scale given the intensity of data requirements. The underlying concern is that misclassifying individuals' exposure may introduce bias or reduced precision in ultimate estimates of the population impacts on health. However, this issue of exposure misclassification has been carefully studied in epidemiological studies, including those of air pollution, where reliance on broad geographic characterizations of exposure is common. In general, the findings from epidemiology suggest that, in theory and in practice, the use of these population exposures estimate likely leads to random error in in the true exposures of individuals and has the effect of dampening the observed effect estimates – that is that they are biased low. There is some evidence that when exposure estimates better approximate personal exposures, that the size of the effect estimates increases. The bottom line is that the large population-based epidemiological studies that form the basis for our understanding of air pollution health effects have not relied on methods like those in Soares et al. Population-weighted exposures have been adopted as the basis for estimating the burden of disease from air pollution in the

Global Burden of Disease project run by the Institute for Health Metrics and Evaluation (Cohen et al., 2017). IHME's methodology is also now used by the World Health Organization.

Revisions in the manuscript:

1. Page 10, line 28-29； page 11, line 1-6: "Soares et al. (2014) built a refined model for evaluating population exposures to ambient air pollution in different microenvironment. In the absence of detailed individual exposure estimates, population-weighted $PM_{2.5}$ concentrations are a better approximation of potential human exposure because they give proportionately greater weight to concentrations in areas where most people live. Population-weighted exposures have been adopted as the basis for estimating the burden of disease from air pollution in the Global Burden of Disease project run by the Institute for Health Metrics and Evaluation (Cohen et al. 2017). IHME's exposure methodology is also now used by the World Health Organization."

16. Page 9, lines 1-2. It seems that the annual estimate is based on two months of actual data. Why not using data for the whole year? This would remove one source of uncertainty from the final results. The lines 9-10 seem to suggest that data for the whole year 2015 was available for the authors.

Response:

Thank you for the question. Due to the limitation of getting the national-scale AIS data for the whole year from the marine-time department, only data in some representative month like January and June are available for our study. Therefore, we used the average values of these two months to estimate annual shipping emissions in whole China. But we have full-year AIS data in Yangtze River Delta (YRD), and the estimates of annual shipping emissions in YRD scale and Shanghai city scale in the manuscript were based on the full-year data. We have clarified the data limitations for national shipping emission estimate in the revised manuscript.

Revisions in the manuscript:

1. *Page 7, line 9-11:* "*<u>Due to limitation of the data source, the national-scale AIS data in this study only covered the representative months of January and June 2015, while the YRD-scale AIS data covered 2015 full year.</u>*"

17. Page 9, lines 18-20. The largest contribution to emissions comes from sources close to the shore. This underlines the importance of including all waterborne traffic sources and consideration of water flow/speed issue. Some discussion of these topics should be included in the manuscript.

Response:

Thank you for this comment. As we responded to "comment 8" and "comment 10", we've added some discussion on the limitation of AIS data and neglecting water flow.

Revisions in the manuscript:

1. Page 19, line 16-25: "*<u>When estimating shipping emission inventory, underestimations of actual emissions may be introduced by missing information. For example, AIS data has a high coverage of coastal vessels, but many inland vessels are not equipped with AIS. Emissions from those inland vessels without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA, and that could introduce some uncertainties for inland river vessels. Also, emissions from fishing boats were probably underestimated because AIS devices on some fishing boats may not be in use. Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions.</u>*"

2. Page 19, line 26-29; page 20, line 1-8: "*<u>In addition, we did not consider the external effects of water flow, wind, and waves when calculating engine power for ships going over the region. This would introduce some uncertainties (Aulinger et al., 2016). According to the previous studies in other areas, these factors may increase fuel consumption of individual vessels by as much as 10% to 20%, while the effects of</u>*

waves on emissions estimations over extensive geographical regions are negligible (Jalkanen et al., 2009; Jalkanen et al., 2012). The downstream of the Yangtze River is located in the geographically plateau region, and the river flow is below 0.5 m/s (Song and Tian, 1997; Xue et al., 2004). For Shanghai, located at the end of mouth of the Yangtze River to the East China Sea with a flat terrain, the river flow is very slow. Given that ships traveling the Yangtze River near Shanghai have speeds over ground (SOG) of about 5-10 knots (3-5 m/s), the relative ratios of water flow to the SOG is within 20%. In our future work, we will fill the gap in the basic ship data and consider the external effects when building the shipping emission inventory.”

18. Page 10, lines 8-10. Authors identify ships as a significant source of VOCs. Have you considered the role of small boats in VOC emissions? The VOC emission levels allowed for boat engines are significantly higher than those of marine diesel engines and there are a lot of small engines in boats.

Response:

Thank you for the question. “The emissions of SO_2, NO_X, and $PM_{2.5}$ from inland-water ships and coastal ships accounted for the majority of primary emissions from all shipping related sources in Shanghai port, ranging from 72% for VOCs to about 99% for SO_2.” Here the proportion is relative to all shipping related sources, which include inland-water ships, coastal ships, container-cargo trucks and port terminal equipment. In this study, the percentage of VOC emissions from ships relative to all pollution sources was not significant, which was 0.3% in YRD region and 0.6% in Shanghai. We have clarified the text and added some discussion on the underestimation of emissions from small boats to the limitations section.

Revisions in the manuscript:

1. Page 11, line 17-20: “*Based on the whole year 2015 AIS data, the annual emissions of SO_2, NO_X, $PM_{2.5}$, and VOC_s from shipping sectors in YRD region were estimated at 2.2×10^5 tons (one third of the value for China), 4.7×10^5 tons, 2.7×10^4 tons, and 1.2×10^4 tons, respectively, which accounted for 7.4%, 11.7%, 1.3%, and 0.3% of the total emissions from all sources in the YRD in 2015.*”

2. Page 12, line 21-23: "The emissions of SO_2, NO_X, $PM_{2.5}$, and VOC_S from inland-water ships and coastal ships accounted for the majority of primary emissions from all shipping related sources in Shanghai port, ranging from 72% for VOCs to about 99% for SO_2. They comprised about 17.4% of SO_2, 24.5% of NO_X, 5.2% of $PM_{2.5}$ and 0.6% of VOC_S emissions from all pollution sources in Shanghai."

3. Page 19, line 16-25: "When estimating shipping emission inventory, underestimations of actual emissions may be introduced by missing information. For example, AIS data has a high coverage of coastal vessels, but many inland vessels are not equipped with AIS. Emissions from those inland vessels without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA, and that could introduce some uncertainties for inland river vessels. Also, emissions from fishing boats were probably underestimated because AIS devices on some fishing boats may not be in use. Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions."

19. Page 12, lines 19-21. If the atmospheric conversion of gaseous SO2 to particulate SO4 takes about a week (reacts with OH), why is the 12 nm distance relevant in this aspect? Surely during one week the gaseous SO2 travels further that 12 nm during that time and it cannot be used as an only explanation why ships further out than 12 nm do not contribute to SO2.

Response:

Thank you for the question. The results showed that shipping within 12 NM was a major contributor to ship-related SO_2 concentrations in core YRD cities, which accounted for at least 78% of the ship-related contribution. Shipping further out than 12 NM accounted for 2% to 17% of the ship-related contribution to SO_2 concentrations in different core YRD cities. Here 12 NM is a reference distance for comparison of different transport range between precursors like SO_2 and aerosol.

Shipping within 12 NM was dominant in ship-related $PM_{2.5}$ concentrations in core YRD cities. However, the contribution from shipping further out 12 NM to $PM_{2.5}$ also substantial, accounting for 17% to 49% of the ship-related contribution (especially busy north-south shipping lanes 24-96 NM, accounting for 12 to 39%). It indicates that the ship-related $PM_{2.5}$ concentrations could also be substantially affected by shipping beyond 12 NM, especially when compared with the SO_2 result. That also implied that the future ECA boundary should consider multiple air pollutants synchronically. This comparison of results could have policy implication and has been clarified in the revised manuscript. In addition, we've expanded the possible reasons why ships further out than 12 NM had much smaller impact on land SO_2 concentrations.

Revisions in the manuscript:

1. Page 15, line 8-21: "Shipping emissions beyond 12 NM had limited contribution to SO_2 concentrations in 16 core YRD cities, implying that the boundary of 12 NM might be suitable for regulating SO_2 emissions. This could also be proved by Schembari et al., (2012), who reported that statistically significant reductions of SO_2 levels (66% to 75%) were found in 3 out of the 4 European harbours, 5 months after the implementation of the EU directive 2005/33/EC that requires all ships at berth or anchorage in European harbours use fuels with a sulfur content of less than 0.1% from January 2010. The quicker chemical reaction and shorter lifetime of SO_2 may explain why ships further out than 12 NM had much smaller impact on land ambient SO_2 concentrations (Collins et al., 2009; Krotkov et al., 2016). SO_2 reacts under tropospheric conditions via both gas-phase processes (with OH) and aqueous-phase processes (with O_3 or H_2O_2) to form sulfate aerosols, and is also removed physically via dry and wet deposition (Seinfeld and Pandis, 2006). The sulfur deposition due to shipping emissions is mainly contributed by the dry depositions (Chen et al.,2019). In the Planet boundary layer (PBL), SO_2 has short lifetimes (less than 1 day during the warm season) and are concentrated near their emission sources (Krotkov et al., 2016)."

2. Page 16, line 4-9: "The results of these YRD analyses suggest that although *ambient ship-related SO_2 concentrations were mainly affected by shipping inland or within 12 NM, expanding China's current DECA to around 100 NM or more would reduce the majority of the impacts of shipping on regional $PM_{2.5}$ pollution. It also implies that the future ECA policy should consider multiple air pollutants including the primary and secondary pollutants synchronically."*

20. Figure 1. The legend text font size should be increased, it is very small reading as it is now. It is especially tough to read the text of the right hand side zoomed images.

Response:

Thank you for the suggestion. The text font size has been increased.

Revisions in the manuscript:

Figure 15

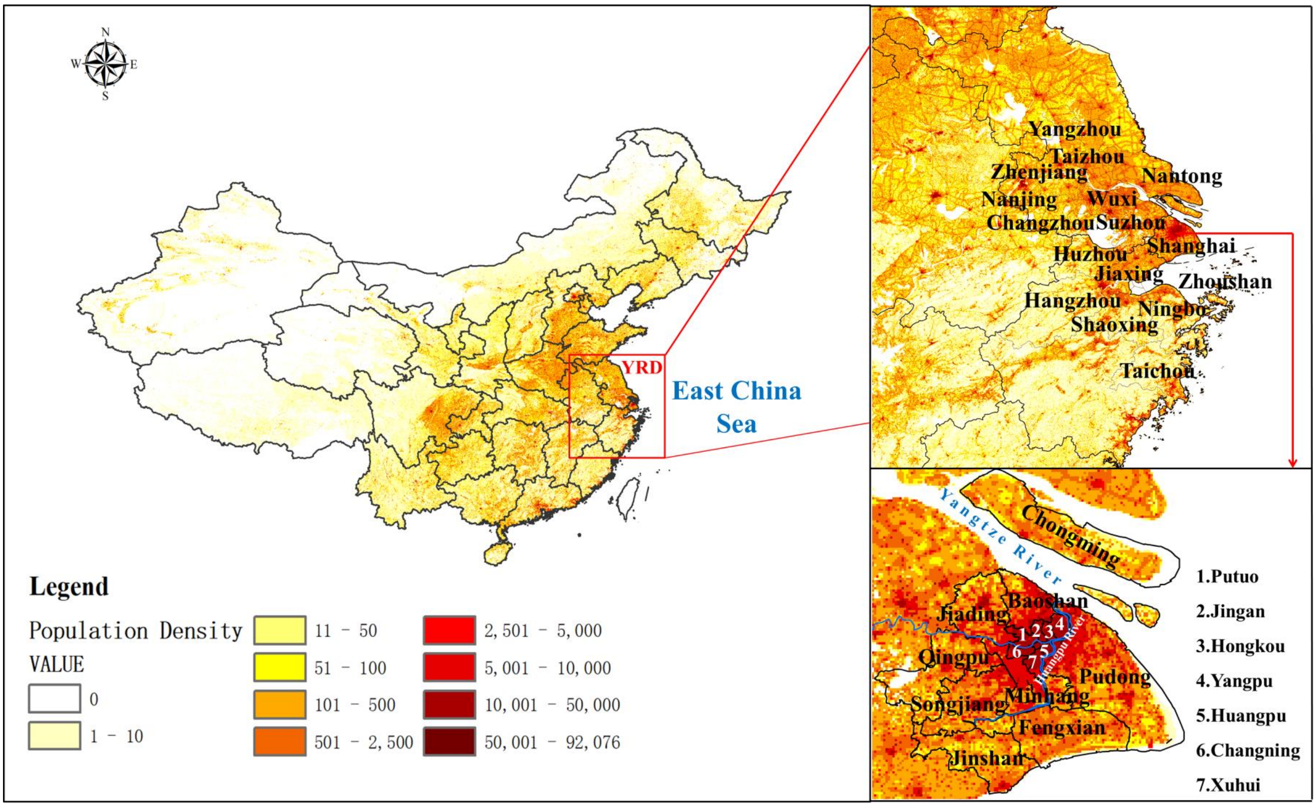


Figure 1. Geographic location of the study area YRD/Shanghai with population density in 2015. 16 core cities in YRD and 16 administrative districts in Shanghai are noted on the map. The smaller administrative districts are labeled with numbers:

Putuo (1), Jingan (2), Hongkou (3), Yangpu (4), Huangpu (5), Changning (6), Xuhui (7).

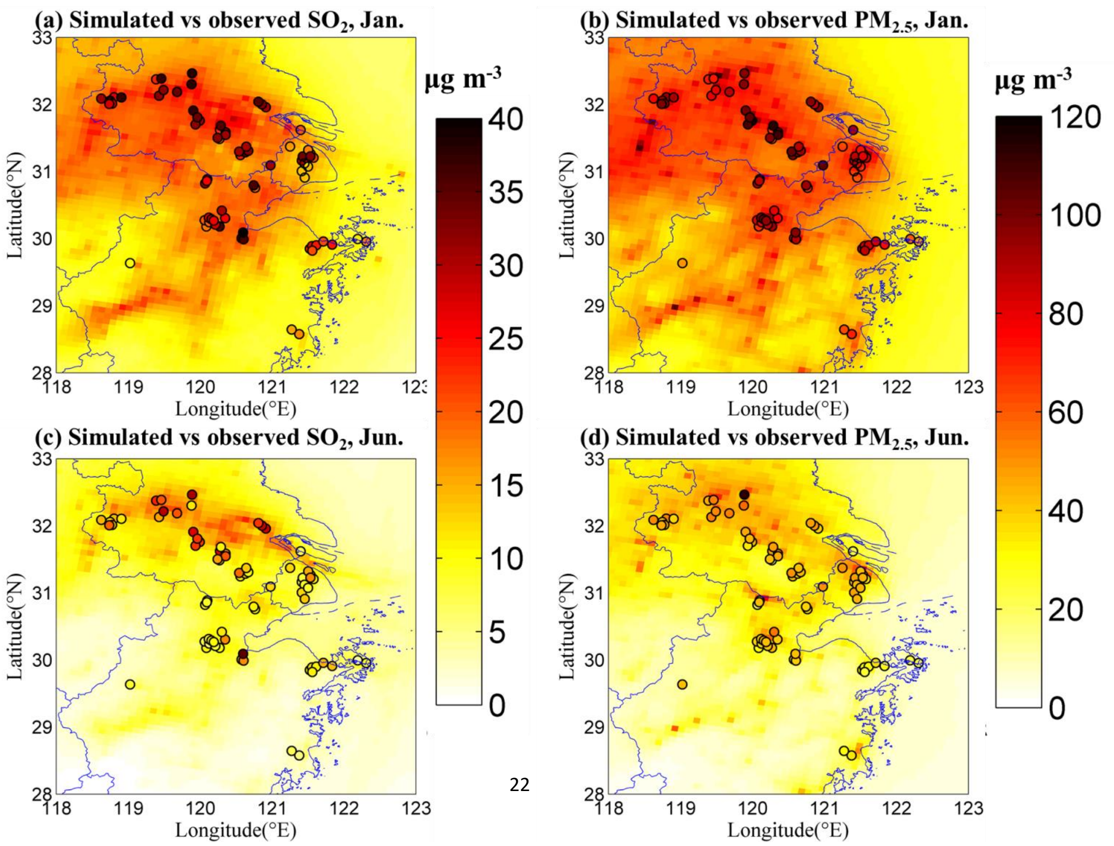
21. Figure 2. This figure is confusing. If the symbols represent measurement values, I cannot see any numerical values linked to the symbols. If the colors correspond to gridded model concentrations, that is fine, but the measured values cannot be determined from these images. Perhaps another form of graphic could be used to provide the comparisons?

Response:

Thank you for this comment. The colors correspond to both gridded model concentrations and measured values (in circle). We have increased the circle size so that the fill color is now more visible. This figure has been moved to supporting information (Figure S2).

Revisions in the manuscript:

Supporting information, Figure S2



<u>Figure S2</u>. The simulated (grid) and observed (circles) SO_2 concentration distribution in YRD region, in January 2015 (a) and June 2015 (c); the simulated (grid) and observed (circles) $PM_{2.5}$ concentration distribution in YRD region, in January 2015 (b) and June 2015 (d)

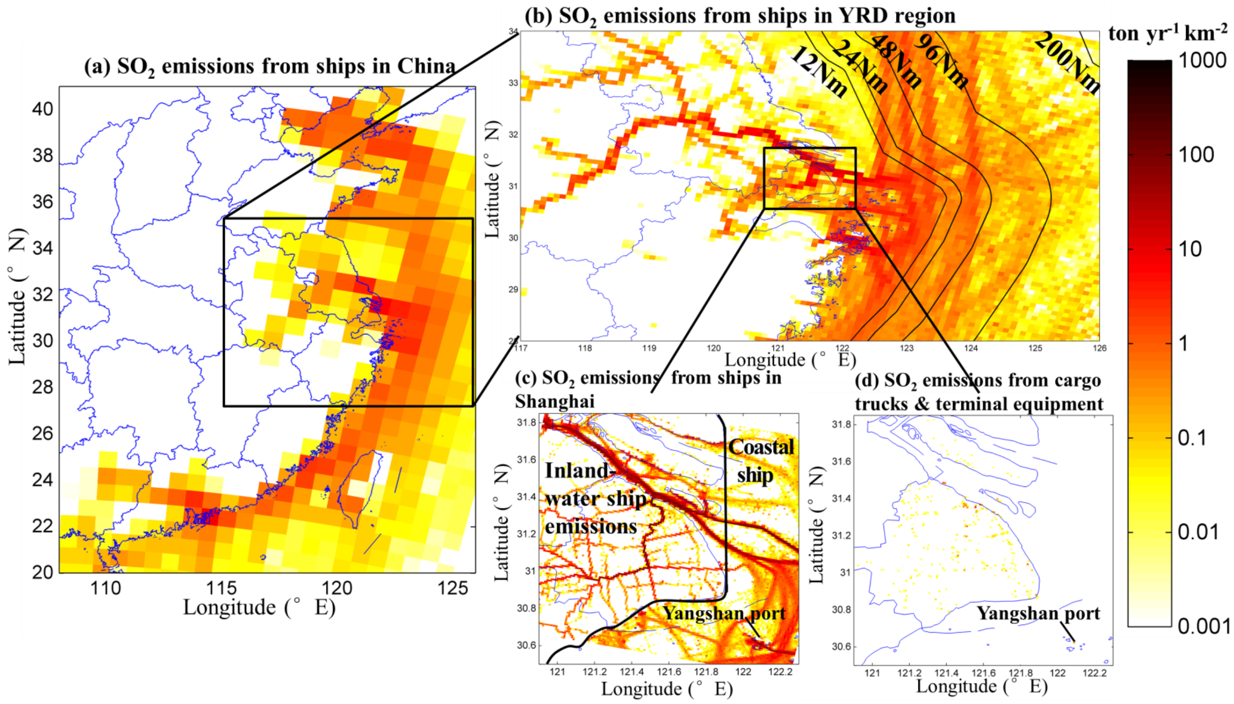
22. Figure 3. The legend texts are very small in this figure, too.

Response:

Thank you for the suggestion. The legend text font size has been increased.

Revisions in the manuscript:

<u>Figure 2</u>



<u>Figure 2</u>. SO_2 emissions in 2015 from (a) shipping traffic in China (the average value of January and June) at resolution of 81km × 81km; (b) ships in different offshore coastal areas (inland-water and within 12 NM, 12-24 NM, 24-48 NM, 48-96 NM and 96-200 NM) in the YRD region, at resolution of 9km × 9km; (c) inland-water ships and coastal ships in Shanghai, at resolution of 1km × 1km; and (d) container-cargo trucks and port terminal equipment in Shanghai, at resolution of 1km × 1km. The

black line in (c) refers to the division line between the inland water and coastal water for Megacity Shanghai defined in this study.

23. Figure 5, I would welcome some discussion why the distance to the shore is relevant in this context. Are the authors trying to see whether it is useful to limit the distance of regulated emissions to a specific value or what is the reasoning of choosing these distance bins?

Response:

Thank you for the question. The distance boundary of current Domestic Emission Control Areas (DECA) in China is 12 NM zone along the coastline. We are assessing the impacts of shipping within 12 NM as well as shipping in offshore coastal areas beyond 12 NM. We referred to ICCT's working paper (Mao et al., 2017) to choose the distance bins (12 NM, 24 NM, 48 NM, 96NM, 200NM) in this study. Among these distance bins, 12 NM is the boundary of current DECA in China and 200 NM is the boundary of ECA designated by IMO. In addition, we considered that shipping at further distances could have a smaller impact on air quality on the land, therefore, the values between 12 NM and 200 NM were doubled in order to make a better comparison among these offshore coastal areas. The results can provide evidence when a specific value is considered for the distance of regulated emissions in future ECA policy in China. We've clarified the reason of choosing the distance bins in the revised manuscript.

Revisions in the manuscript:

1. Page 5, line 30; page 6, 1-2: "The results of this study could be informative to <u>the consideration of the distance of regulated emissions</u> in the design of future emissions control areas for shipping <u>in YRD</u>"

2. Page 6, line 9-11: "Then, we used WRF-CMAQ model to evaluate the impacts on air quality from shipping emissions in different offshore coastal areas (within 12 NM including inland waters, 12-24 NM, 24-48 NM, 48-96 NM, and 96-200 NM) in the YRD region. <u>We referred to ICCT's working paper (Mao et al., 2017) to choose the</u>

distance bins between 12 NM (the boundary of current China's DECA) and 200 NM (the boundary of ECA designated by IMO) in this study."

24. Figure 9. Texts are too small, especially in the two top images.

Response:

Thank you for the suggestion. The text font size has been increased.

Revisions in the manuscript:

Figure 8

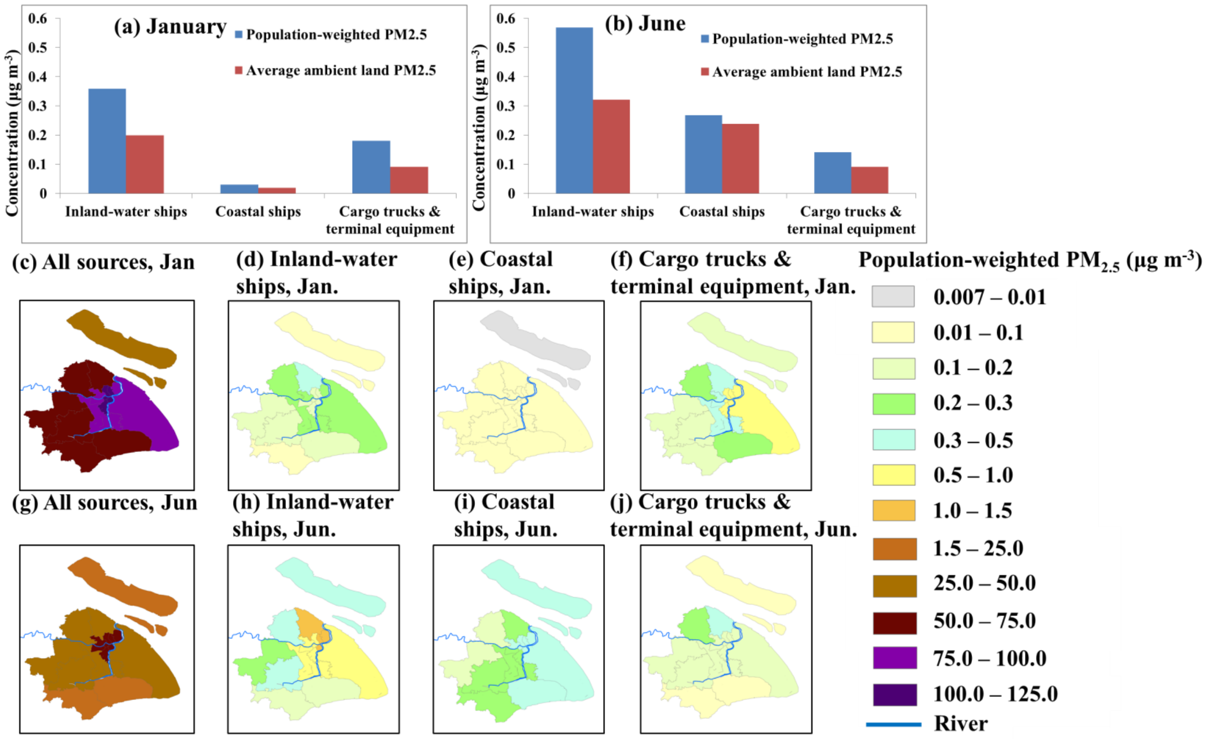


Figure 8. Population-weighted $PM_{2.5}$ and average $PM_{2.5}$ caused by different ship-related sources in Shanghai, in January(a) and in June (b); population-weighted $PM_{2.5}$ caused by all pollution sources (c, g), inland-water ships (d, h), coastal ships (e, i) and container-cargo trucks and port terminal equipment (f, j) in 16 districts in Shanghai, in January 2015(c-f) and June 2015 (g-j)

25. Table 1. Are these daily, monthly or annual values? There is no indication of the timeline here? This data does not tell me very much of how well the model is able to

capture the temporal variability of pollution peaks. Could a line graph be used here instead? This would help to see how well the model is able to capture the air concentrations.

Response:

Thank you for this comment. Observed data (Obs.) and simulated data (Sim.) for each city are the average of monthly values of January and June case. The statistical metrics of NMB, NME, RMSE and *r* were calculated based on the daily-average observed and simulated data. We've clarified the timeline in the caption of Table 1.

We've made line graphs of temporal variability. However, if we put all the line graphs in the manuscript, it would be too many (32 line graphs for 16 cities and two pollutants). So we chose line graphs of four representative cities (two coastal cities and two inland cites) to be presented in the supporting information.

Revisions in the manuscript:

1. Table 1, caption

"Table 1 Statistical metrics of the model evaluation. Observed data (Obs.) and simulated data (Sim.) for each city are the average of monthly values of January and June case. NMB, NME, RMSE and r were calculated based on the daily-average observed and simulated data."

2. Supporting information, Figure S3

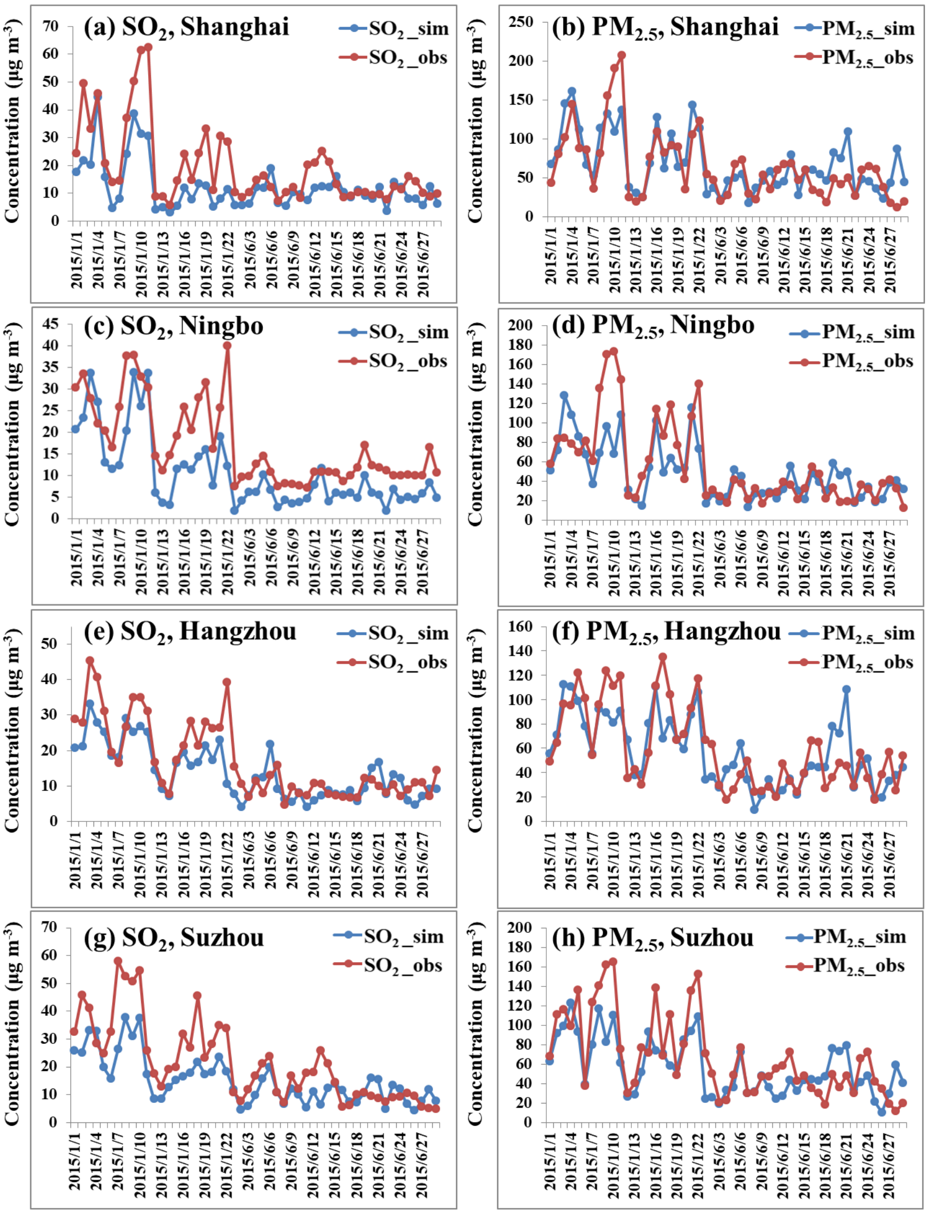


Figure S3 Daily variability of simulated (sim.) and observed (obs.) SO_2 concentrations (a, c, e, g) and $PM_{2.5}$ concentrations (b, d, f, h) in four representative cities, including two coastal cities – Shanghai (a, b) and Ningbo (c, d), and two inland cites – Hangzhou (e, f) and Suzhou (g, h).

26. Table 2. No units are given?

Response:

Thank you for the question. The units were given in the left column which may not be very obvious. We've clarified the units in the title.

Revisions in the manuscript:

Table 2. Primary emissions (ton/yr), emission share in all shipping emission (%) and emissions density (ton/yr/km^2) from shipping at different boundaries in YRD region[a] in 2015

	Pollutants	Within 12 NM	12-24 NM	24-48 NM	48-96 NM	96-200 NM
Shipping	SO_2	1.3×10^5	1.4×10^4	2.5×10^4	3.2×10^4	1.3×10^4
emission	NO_x	3.6×10^5	2.0×10^4	3.5×10^4	4.5×10^4	1.8×10^4
inventory	$PM_{2.5}$	1.3×10^4	2.4×10^3	4.5×10^3	5.4×10^3	1.5×10^3
(ton/yr)	VOC_s	7.9×10^3	8.3×10^2	1.3×10^3	1.5×10^3	3.0×10^2
Emission	SO_2	61.4	6.4	11.4	14.9	5.8
share in all	NO_x	75.0	4.1	7.4	9.6	3.9
shipping	$PM_{2.5}$	48.4	9.0	16.9	20.2	5.5
emission (%)	VOC_s	66.6	7.0	11.2	12.6	2.6
Emission	SO_2	0.66	0.54	0.49	0.33	0.06
density	NO_x	1.74	0.86	0.77	0.51	0.08
(ton/yr/km^2)	$PM_{2.5}$	0.08	0.06	0.06	0.04	0.01
	VOC	0.05	0.02	0.01	0.01	0.001

a. domain 3

27. Table 3. No units given?

Response:

Thank you for the question. The units were given in the left column which may not be very obvious. We've clarified the units in the title.

Revisions in the manuscript:

Table 3. Primary emissions (ton/yr) and emission share in all pollution sources (%) from different-type ship-related source in Shanghai[a] in 2015

	Ship-related source	*SO_2*	*NO_x*	*$PM_{2.5}$*	*VOC*
Emission inventory (ton/yr)	*Inland-water ships[b]*	*3.3×10^4*	*9.2×10^4*	*0.40×10^4*	*0.27×10^4*
	Coastal ships[c]	*1.6×10^4*	*2.9×10^4*	*0.18×10^4*	*0.067×10^4*
	Container-cargo trucks	*0.0*	*1.8×10^4*	*0.064×10^4*	*0.11×10^4*
	Port terminal equipment[d]	*0.0021×10^4*	*0.18×10^4*	*0.0057×10^4*	*0.022×10^4*
Emission share in all pollution sources in Shanghai (%)	*Inland-water ships*	*11.8*	*18.7*	*3.6*	*0.5*
	Coastal ships	*5.6*	*5.8*	*1.6*	*0.1*
	Container-cargo trucks	*0.0*	*3.7*	*0.6*	*0.2*
	Port terminal equipment	*0.01*	*0.36*	*0.05*	*0.04*

a. domain 4

b. defined as ships operate in both the outer port and in the inner river region of Shanghai Port, which include Yangtze River, Huangpu River and other river ways in Shanghai

c. includes China coastal and international ships

d. includes cranes and forklifts used for internal transport

Supplementary material, S1

28. The authors seem to apply the Starcrest methodology in their emission modeling.

Page 2, text under Eq (1). Maximum speed and design speed of ships are two different things and IHS data often mentions economic speed. Which was one was actually used in the analysis?

Response:

Thank you for the question. Maximum speed was used in the analysis. We've corrected this sentence into "where ActSpeed is the actual speed when ship is cruising and MaxSpeed is the maximum speed for the ship".

Revisions in the manuscript:

1. Supporting information, Page 2, line 8-9: "where ActSpeed is the actual speed when ship is cruising and MaxSpeed is the maximum speed for the ship"

29. Page 2, near Eq (4), aux boiler use. Did the authors consider the exhaust boilers at all in this regard? Also, the installed boiler capacity is difficult to determine from ship databases, because this field is not provided. I would like to know where the installed boiler data comes from.

Response:

Thank you for the question. We did not consider the exhaust boilers because limited information on boilers could be found in Lloyd's database and China Classification Society (CCS) database. We've add some discussion about the underestimation of boiler emissions in the new section on limitations.

Revisions in the manuscript:

1. Supporting information, page 2, line 28-30: "However, auxiliary boiler emissions were not considered in this study because limited auxiliary boiler information exists in the Lloyd's register and Chinese Classification Society (CCS) database."

2. Page 19, line 23-25: "Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions."

30. Page 2, last paragraph. Authors make reference to Lloyds, 2009 which is not listed in the bibliography provided for S1. Also, why refer to data from 2009 if the AIS data is for 2015. How were the ships built during 2009-2015 treated?

Response:

Thank you for the question. In fact, the 2015 Lloyd's register was used in this study. We have corrected this mistake and also listed the reference in the bibliography for S1.

Revisions in the manuscript:

1. Supporting information, page 2, line 35-36: "For ships available in Lloyd's register (now IHS-Fairplay) (Lloyd's register, 2015)"

2. Supporting information, References: "Lloyd's register (IHS Fairplay). 2015"

31. Page 2, last paragraph. Authors assume all inland waterway vessels to have 7000 kW engine? No effort was made to identify these vessels and use proper description of installed power?

Response:

Thank you for the question. We did not assume all inland waterway vessels to have 7000 kW engine. Information of some domestic ships is available in Chinese Classification Society (CCS) database, and description of installed power was used according to the information in the database. But for those domestic ships which are not available in Lloyd's register and CCS database, their main engine power was assumed to be 7000 kw by default, which was close to the domestic ships from the CCS database (with main engine power mainly ranging from 4000 kw to 6000 kw), and below the East China Sea-going ships in Lloyd's register (with main engine power mainly ranging from 11000 kw to 14000 kw). We've clarified this in the revised manuscript. Also we will fill the gap in the basic ship data in future work.

Revisions in the manuscript:

1. Supporting information, page 3, line 2-6: "Information of some domestic ships is available in CCS database, but for those ships unavailable in the database, the main engine power was assumed to be 7000 kw by default, which was close to the domestic ships from the CCS database (with main engine power mainly ranging from 4000 kw to 6000 kw) and below the East China Sea-going ships in Lloyd's register (with main engine power mainly ranging from 11000 kw to 14000 kw)"

32. Page 3, S2, first paragraph. "Table S1 lists emission factors used in the present study". This is not true and the emission factor table is missing.

Response:

Thank you for the correction. Since the emission factor table was already in our previous work (Fan et al. 2016), we did not put the table in the supporting information in this study repeatedly. We are sorry for the written mistake. We've modified this sentence into "Emission factors used in the present study were listed in Fan et al. (2016)."

Revisions in the manuscript:

1. Supporting information, page 3, line 31-32 "Emission factors used in the present study were listed in Fan et al. (2016)."

2. Supporting information, References: "Fan, Q., Zhang, Y., Ma, W., Ma, H., Feng, J., Yu, Q., Yang, X., Ng, S. K., Fu, Q., and Chen, L.: Spatial and Seasonal Dynamics of Ship Emissions over the Yangtze River Delta and East China Sea and Their Potential Environmental Influence, Environ. Sci. Technol., 50, 1322-1329, 10.1021/acs.est.5b03965, 2016."

33. Page 3, second paragraph. Add reference ICF, 2009

Response:

Thank you for the correction. The reference has been added.

Revisions in the manuscript:

1. Supporting information, References: "ICF International: Current methodologies in preparing mobile source port-related emission inventories, 2009."

34. Page 3, second paragraph. OC and EC low load adjustment factors were treated the same way as PM. This is contrary to the behavior of EC and EC as a function of engine load. Authors might want to check the ICCT report "Black Carbon Emissions and Fuel Use in Global Shipping, 2015", Oct 2017 for low load behavior of carbon fraction.

Response:

Thank you for this comment. In this study, emission factors were adjusted for loads below 20 % using values from studies conducted in other countries (ICF International, 2009; Starcrest Consulting Group, 2009). Because OC and EC low load adjustment factors were not available in these studies, they were assigned the same as PM. In ICCT's report "Black Carbon Emissions and Fuel Use in Global Shipping, 2015" (ICCT, 2017), the authors mentioned that "Emission factors tend to increase at low loads. Low load adjustment factors from the Third IMO GHG Study 2014 were applied when estimated main engine load fell below 20% for all pollutants except BC, which is not estimated in the IMO study. In this case, BC EFs are determined from power curves described in the previous section, which already account for changes in BC EFs as a function of engine load." From the power curves of BC EF (shown in the Figure R1 below), it indicated that the BC EFs increase significantly at low loads, especially below 20%. The unit of EF given in ICCT's report (ICCT, 2017) is g/kg fuel, while the unit of EF given in this study is g/kWh, which is hard to make direct comparisons. So we've estimated the proportion of BC emissions in PM emissions (BC/PM). In this study, BC/PM was 0.029, a bit lower than the value 0.045 in ICCT's report. Petzold et al. (2004) measured a BC fraction of 2% of the total particle mass for an engine load of 100%. Erying et al. (2005) estimated shipping emissions based on fuel consumption, and reported 0.05 Tg of BC and 1.67 Tg of PM_{10}, and BC/PM was around 0.03. Therefore, the ratio of BC to PM in this study is within a reasonable range. We've added some discussion about the uncertainty brought by the selection of low load adjustment factors in the supporting information.

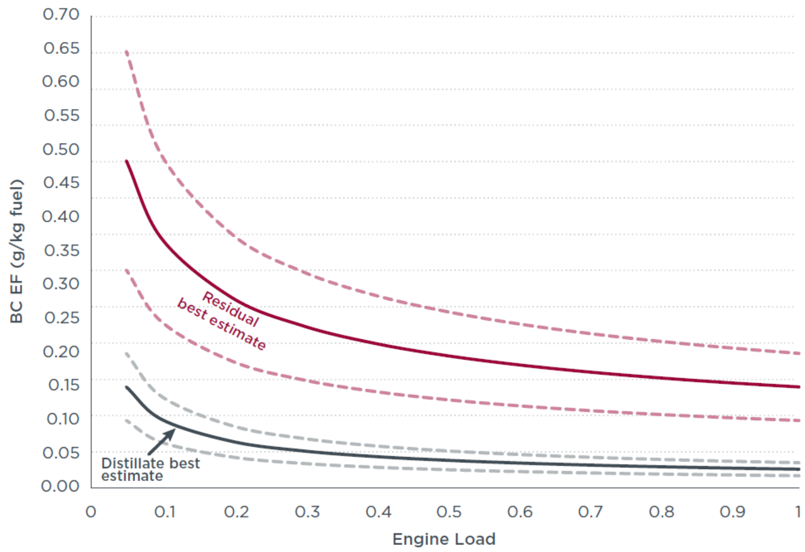


Figure R1. Black carbon emission factors for 2-stroke engines by fuel type (ICCT, 2017)

Revisions in the manuscript:

1. Supporting information, page 3, line 34-39: "Because adjustment multipliers were not available for organic carbon (OC) and elemental carbon (EC), these pollutants were assigned the same low load adjustment multiplier (LLAM) as PM in the present *study, <u>which may introduce uncertainties. In this study, the ratio of BC emissions to PM emissions (BC/PM) was around 2.9%, which falls within the range of 2% to 4.5% in other studies (Comer et al., 2017; Erying et al., 2005; Petzold et al., 2004).</u>"*

References:

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Soares, J., Kousa, A., Kukkonen, J., Matilainen, L., Kangas, L., Kauhaniemi, M., Riikonen, K., Jalkanen, J. P., Rasila, T., Hänninen, O., Koskentalo, T., Aarnio, M., Hendriks, C., and Karppinen, A.: Refinement of a model for evaluating the population exposure in an urban area, Geosci. Model Dev., 7, 1855-1872, 10.5194/gmd-7-1855-2014, 2014

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Response to Referee's Comments #2

1. General comments: This study presented the importance of geographical locations of ship emissions to the environmental and human health effects. The manuscript has been well written and organized. Take the YRD region– one of the busiest port cluster in the world as the example, this study result is helpful to understand the meaningful points of future ECA policy. The authors should explicit the key implication through the paper, including the abstract, result and conclusion part.

Response:

Thank you for the comments and the suggestions. We have expanded on the key implications of these research results for potential ECA regulations throughout the manuscript.

Revisions in the manuscript:

1. Page 2, line 6-8: "in particular, in the YRD region, expanding the boundary of 12 NM in China's current DECA policy to around 100 NM would include most of the shipping emissions affecting air pollutant exposures, and stricter fuel standards could be considered for the ships on inland rivers and other waterways close to residential regions."

2. Page 5, lines 6-15: "In China, a few studies reported the contribution to air pollution from shipping in different offshore coastal areas or different-type ship-related sources. For example, Mao et al. (2017) estimated primary emissions from OGVs at different boundaries in the PRD region, and concluded that further expansion of emission control area to 100 NM would provide even greater benefits. However, the impacts of shipping emissions at varying distances from shore on air quality and potential human exposure, which are important when considering ECA policy, have not been rigorously studied. Mao and Rutherford (2018) studied NO_x emissions from three categories of merchant vessels—OGVs, coastal vessels (CVs) and river vessels (RVs) in China's coastal region. But less attention was paid to the impacts of inland waterway traffic and port-related sources like container-cargo trucks and terminal port equipment on air quality and potential human exposure."

3. Page 5, line 30; page 6, line 1-3: "The results of this study could be informative to *the consideration of the distance of regulated emissions in the design of future emissions control areas for shipping in YRD, or regulations on the sulfur content of fuels for different-type ship-related sources in Shanghai."*

4. Page 16, line 6-9: "The results of these YRD analyses suggest that although ambient ship-related SO_2 concentrations were mainly affected by shipping inland or within 12 NM, expanding China's current DECA to around 100 NM or more would reduce the majority of the impacts of shipping on regional $PM_{2.5}$ pollution. It also implies that the future ECA policy should consider multiple air pollutants including the primary and secondary pollutants synchronically."

5. Page 19, line 10-12: "The results of the analyses of different-type shipping-related sources indicated that ship-related sources close to densely-populated areas contribute substantially to population exposures to air pollution."

6. Page 20, line 28-29; page 21, line 1-4: "For example, policymakers could consider whether to expand China's current DECA boundary of 12 NM to around 100 NM or more to reduce the majority of shipping impacts on air pollution concentrations and exposure. It will be helpful to improve the local air quality and reduce human exposures in densely populated areas by developing more stringent regulations on the fuel quality for ships entering inland rivers or other waterways close to residential regions."

The details should be improved:

2. Page 6-7, 2.2.2 Non-shipping emission inventories part. For the national scale domain and regional scale domain, several sets emission data has been used. The authors should make clearer how they merge the emission together. How did they use 2015 national emission database to make a regional 27 km × 27 km resolution that included 5 pollutants? Did they use spatial interpolation method? Which year are the IIASA data for CO and NH3?

Response:

Thank you for the question. The 2015 national emission database (including PM_{10}, $PM_{2.5}$, SO_2, NO_x and VOCs) at a 27 km × 27 km resolution (Zhao et al., 2018) and

2015 IIASA database at a 0.5°× 0.5°resolution (CO and NH_3) (Stohl et al., 2015) was allocated to domain 1 (81 km × 81 km) and domain 2 (27 km × 27 km) by spatial interpolation in Arcgis 10.2. We have clarified the method and the year of IIASA data in the revised manuscript.

Revisions in the manuscript:

1. Page 8, line 19-21: "supplemental emission data on these pollutants in 2015 were obtained from the International Institute for Applied Systems Analysis (IIASA) database (at a 0.5°× 0.5°resolution) (Stohl et al., 2015)."

2. Page 8, line 29-30: "National and local emission data were allocated to simulation grids by spatial interpolation in ArcGIS 10.2 (ESRI, 2013)."

3. Page7, line 15-16: "The initial and boundary conditions for meteorology were generated from the Chinese National Centers for Environmental Prediction (NCEP) Final Analysis (FNL)", here the authors should confirm the NCEP FNL data source.

Response:

Thank you for pointing out this. We are sorry for the written mistake. The data source should be "National Centers for Environmental Prediction (NCEP) Final Analysis (FNL)" and it has been corrected in the revised manuscript.

Revisions in the manuscript:

1. Page 9, line 6-8: "The initial and boundary conditions for meteorology were generated from the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) (NCEP, 2000)"

4. Page 9, line 12-17: The authors compared the result of YRD shipping emission with Fan et al.'s and Chen et al.'s studies. The authors quoted Liu et al. (2018) to compare the proportion of YRD shipping emissions in whole China. However, Liu et al. (2018) also reported YRD shipping emissions. Why not compare the result with the values in Liu et al. (2018) as well?

Response:

Thank you for the suggestion. We've added the comparison with the 2013 YRD shipping emission estimates in Fu et al. (2017). This paper is from the same research group as Liu et al. (2018), but reports more pollutants than in Liu et al. (2018). In addition, the comparison with the results in Fan et al. (2016) has been removed because the values were for the year 2010, much earlier than the baseline year 2015 in this study.

Revisions in the manuscript:

1. Page 11, line 20-24: "The emission estimates of SO_2, NOx and $PM_{2.5}$ were slightly lower than Chen et al.'s estimates for 2014 year due to the different temporal or spatial statistical scope (Chen et al., 2019; Fu et al., 2017)."

2. References: "Fu, M., Liu, H., Jin, X., and He, K.: National- to port-level inventories of shipping emissions in China, Environ. Res. Lett., 12, 114024, 10.1088/1748-9326/aa897a, 2017."

5. Page 10, line 12-16: The authors quoted Fu et al. (2012), which used 2010 vessel call data to estimate shipping emissions. I suggest authors reviewed recent studies using AIS data to make comparisons in Shanghai port.

Response:

Thank you for the suggestion. We've reviewed the results in Fu et al. (2017) which reported 2013 shipping emissions in Shanghai Port using AIS data. We've added some discussion on the comparison between the values in this study and in Fu et al.'s study.

Revisions in the manuscript:

1. Page 12, line 26-29; page 13, line 1-3: "Emissions estimates from this study fall within the range of estimates from other studies (Fu et al., 2012; Fu et al., 2017). On the basis of shipping visa data, Fu et al. (2012) determined that the total amounts of SO_2, NO_X, and $PM_{2.5}$ in the vicinity of Shanghai port in 2010 were 3.5×10^4 ton/yr, 4.7×10^4 ton/yr, and 3.7×10^3 ton/yr, respectively, substantially lower than estimates in this study. Using AIS data, Fu et al. (2017) reported 5×10^4 tons of SO_2 and 7×10^4 tons of NOx from shipping in Shanghai port in 2013, close or a bit lower than the results in this study."

6. Page 12, line 6-15: The contribution to SO2 from ships in different coastal areas was not discussed in this paragraph. But in the following paragraph, the authors discussed cumulative contributions from ships at different distance to both SO2 and PM2.5. It shows no consistency when authors discussed SO2 results throughout the section 3.2.2.

Response:

Thank you for this comment. We've supplemented the data of average and peak contribution to SO_2 from ships in different coastal areas in Table S4. Also, we've added some discussion in the revised manuscript.

Revisions in the manuscript:

1. Page 14, line 19-22: "The average and peak contributions from the shipping emissions in specific offshore coastal areas to the ambient SO_2 and $PM_{2.5}$ concentrations on shore for the two months are listed in Table S4. Shipping emissions beyond 12 NM had a much smaller impact on ambient SO_2, which average contributions were below 0.01 $\mu g/m^3$ and peak contributions were below 0.06 $\mu g/m^3$ (Table S4)."

2. Table S4 Average and peak contributions from ship emissions in different offshore coastal areas to the ambient SO_2 and $PM_{2.5}$ concentrations in January and June

Offshore distance	Average contribution ($\mu g/m^3$)				Maximum contribution ($\mu g/m^3$)			
	SO_2		$PM_{2.5}$		SO_2		$PM_{2.5}$	
	January	June	January	June	January	June	January	June
Inland and within 12 NM	0.52	0.70	0.24	0.56	6.00	8.79	1.62	4.02
12-24 NM	0.005	0.007	0.01	0.04	0.03	0.05	0.05	0.20
24-48 NM	0.01	0.009	0.04	0.07	0.06	0.05	0.11	0.34
48-96 NM	0.02	0.008	0.07	0.07	0.05	0.03	0.14	0.30
96-200 NM	0.00	0.001	0.003	0.01	0.004	0.003	0.02	0.05

7. Page 14, line 1-6: The authors discussed the population-weighted PM2.5 from both

shipping source and all pollution sources. Then, what's the proportion of population-weighted PM2.5 from the shipping source among all pollution sources? I suggest some discussion here.

Response:

Thank you for the suggestion. We have added the proportion of population-weighted $PM_{2.5}$ among all pollution sources along with some discussion.

Revisions in the manuscript:

1. Page 17, line 12-16: "Thus, population-weighted $PM_{2.5}$ concentrations from shipping sources accounted for 0.9% to 15.5% of the population-weighted $PM_{2.5}$ concentrations from all pollution sources in June, larger than the contributions of 0.2% to 1.6% in January, which was attribute to higher shipping-related population-weighted $PM_{2.5}$ concentrations in June and higher population-weighted $PM_{2.5}$ concentrations from all pollution sources in January."

8. Page 15, line 25: The uncertainty analysis is lacked in the section of result and discussion. The uncertainties of shipping emission inventories should be discussed here.

Response:

Thank you for pointing out this. We've added section 3.4 Limitations where we discuss the uncertainties associated with our shipping emission inventories.

Revisions in the manuscript:

1. Page 19, line 14-29; page 20, line 1-8:

*"**3.4 Limitations and uncertainties***

Limitations in the study were mainly related to some missing information in database and assumptions during estimation of shipping emissions. When estimating shipping emission inventory, underestimations of actual emissions may be introduced by missing information. For example, AIS data has a high coverage of coastal vessels, but many inland vessels are not equipped with AIS. Emissions from those inland vessels without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA, and that could introduce *some uncertainties for inland river vessels. Also, emissions from fishing boats were probably underestimated because AIS devices on some fishing boats may not be in use. Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions.*

In addition, we did not consider the external effects of water flow, wind, and waves when calculating engine power for ships going over the region. This would introduce some uncertainties (Aulinger et al., 2016). According to the previous studies in other areas, these factors may increase fuel consumption of individual vessels by as much as 10% to 20%, while the effects of waves on emissions estimations over extensive geographical regions are negligible (Jalkanen et al., 2009; Jalkanen et al., 2012). The downstream of the Yangtze River is located in the geographically plateau region, and the river flow is below 0.5 m/s (Song and Tian, 1997; Xue et al., 2004). For Shanghai, located at the end of mouth of the Yangtze River to the East China Sea with a flat terrain, the river flow is very slow. Given that ships traveling the Yangtze River near Shanghai have speeds over ground (SOG) of about 5-10 knots (3-5 m/s), the relative ratios of water flow to the SOG is within 20%. In our future work, we will fill the gap in the basic ship data and consider the external effects when building the shipping emission inventory."

Reference:

Fu, M., Liu, H., Jin, X., and He, K.: National- to port-level inventories of shipping emissions in China, Environ. Res. Lett., 12, 114024, 10.1088/1748-9326/aa897a, 2017.

Liu, H., Meng, Z. H., Shang, Y., Lv, Z. F., Jin, X. X., Fu, M. L., and He, K. B.: Shipping emission forecasts and cost-benefit analysis of China ports and key regions' control, Environ. Pollut., 236, 49-59, 10.1016/j.envpol.2018.01.018, 2018b.

Stohl, A., Aamaas, B., Amann, M., Baker, L. H., Bellouin, N., Berntsen, T. K., Boucher, O., Cherian, R., Collins, W., Daskalakis, N., Dusinska, M., Eckhardt, S., Fuglestvedt, J. S., Harju, M., Heyes, C., Hodnebrog, Ø., Hao, J., Im, U., Kanakidou, M., Klimont, Z., Kupiainen, K., Law, K. S., Lund, M. T., Maas, R., MacIntosh, C. R., Myhre, G., Myriokefalitakis, S., Olivié, D., Quaas, J., Quennehen, B., Raut, J.-C., Rumbold, S. T., Samset, B. H., Schulz, M., Seland, Ø., Shine, K. P., Skeie, R. B., Wang, S., Yttri, K. E., and Zhu, T: Evaluating the climate and air quality impacts of short-lived pollutants, Atmos. Chem. Phys. 15, 10529–10566, https://doi.org/10.5194/acp-15-10529-2015, 2015.

Zhao, B., Zheng, H., Wang, S., Smith, K. R., Lu, X., Aunan, K., Gu, Y., Wang, Y., Ding, D., Xing, J., Fu, X., Yang, X., Liou, K. N., and Hao, J.: Change in household fuels dominates the decrease in PM2.5 exposure and premature mortality in China in 2005-2015, P. Natl. Acad. Sci. USA, 115, 12401-12406, 10.1073/pnas.1812955115, 2018

List of Revisions in Manuscript

1. *Page 1, line 26-31: "The goal of this study was to estimate the contributions of shipping to regional emissions, air quality, and population exposure and to characterize the importance of the geographic spatiality of shipping lanes and different-type shipping-related sources for the baseline year 2015, prior to the implementation of China's Domestic Emission Control Areas (DECAs) in 2016."*
2. *Page 2, line 5-8: "in particular, in the YRD region, expanding the boundary of 12 NM in China's current DECA policy to around 100 NM would include most of the shipping emissions affecting air pollutant exposure, and stricter fuel standards could be considered for the ships on inland rivers and other waterways close to residential regions."*
3. *Page 2, line 19-24: "Globally, about 50,000 to 90,000 cardiopulmonary diseases and lung cancer deaths were attributable to exposure to particulate matter emitted from shipping in 2010 and 2012, respectively (Corbett et al., 2007;Partanen et al., 2013;Winebrake et al., 2009), and 403,300 premature mortalities per year due to shipping are predicted in 2020 under business-as-usual (BAU) assumptions (Sofiev et al., 2018)."*
4. *Page 4, line 4-5: "The DECA implementation timeline specified that qualified ports are encouraged to be in compliance since April 1, 2016"*
5. *Page 4, line 25-28: "Primary ship-emitted particles measured by an aerosol time-of-flight mass spectrometer were typically 1.0 to 10.0 % of the measured particle number concentration, with the contribution rising to as high as 50.0 % in spring and summer (Liu et al. 2016b)."*
6. *Page 4, line 29-30: "In Guangzhou and Zhuhai, shipping emissions were among the top contributors to $PM_{2.5}$ and accounted for greater than 17% of $PM_{2.5}$ mass concentrations (Tao et al. 2016)."*
7. *Page 5, line 6-15: "In China, a few studies reported the contribution to air pollution from shipping in different offshore coastal areas or different-type ship-related sources. For example, Mao et al. (2017) estimated primary emissions from OGVs at different boundaries in the PRD region, and concluded that further*

expansion of emission control area to 100 NM would provide even greater benefits. However, the impacts of shipping emissions at varying distances from shore on air quality and potential human exposure, which are important when considering ECA policy, have not been rigorously studied. Mao and Rutherford (2018) studied NO_x emissions from three categories of merchant vessels—OGVs, coastal vessels (CVs) and river vessels (RVs) in China's coastal region. But less attention was paid to the impacts of inland waterway traffic and port-related sources like container-cargo trucks and terminal port equipment on air quality and potential human exposure."

8. Page 5, line 16: "To fill this gap"

9. Page 5, line 18-28; page 6, line 1-2: "We modeled shipping emissions in different offshore areas in the YRD region and emissions from different-type ship-related sources in Shanghai city for each month of the year. To identify which offshore areas in the YRD region and which ship-related sources in Shanghai contributed the most ambient air pollution, and human population exposure, we modeled the impacts of shipping emissions in different offshore areas (within 12 NM including inland waters, 12-24 NM, 24-48 NM, 48-96 NM, and 96-200 NM) in the YRD region as well as coastal ships, inland-water ships, and container-cargo trucks and port terminal equipment in and near the port areas under the jurisdiction of Shanghai MSA in two representative months (January and June).The results of this study could be informative to the consideration of the distance of regulated emissions in the design of future emissions control areas for shipping in YRD, or regulations on the sulfur content of fuels for different-type ship-related sources in Shanghai."

10. Page 6, line 9-11: "We referred to ICCT's working paper (Mao et al., 2017) to choose the distance bins between 12 NM (the boundary of current China's DECA) and 200 NM (the boundary of ECA designated by IMO) in this study."

11. Page 6, line 21-22: "Domain 1 covers the whole China."

12. Page 7, line 2-3: "Higher shipping impacts were expected in June because prevailing winds from the summer monsoon are directed from the ocean to the shore."

*13. Page 7, line 5: "***2.2 Emission inventories***"*

14. Page 7, line 7-12: "In this study, emission inventories were constructed based primarily on automatic identification system (AIS) data for shipping traffic activity in China, YRD, and Shanghai geographic domains. Due to limitation of the data source, the national-scale AIS data in this study only covered the representative months of January and June 2015, while the YRD-scale AIS data covered 2015 full year. AIS data includes international ships, coastal ships, and inland-water ships, but some river ships could be not covered by AIS data."

15. Page 7, line 12-16: "Emissions from ships entering the geographic domains for YRD or Shanghai were calculated using the AIS-based model developed by Fan et al. (Fan et al., 2016), and monthly shipping emissions for January and June were used in the air quality model to capture the seasonal variation to expect more accurately than annual shipping emissions with no monthly variations."

16. Page 7, line 19: "supporting information"

17. Page 7, line 23: "Lloyd's register *(now IHS-Fairplay)* (Lloyds,2015)*"*

18. Page 7, line 24-26: "Assumptions regarding the fuel types, sulfur contents and engine types, and sources of emission factors, low load adjustment multipliers, and control factors are provided in section S.2 of the supporting information."

19. Page 8, line 17: "(Zhao et al., 2018)"

20. Page 8, line 17-26: "Since the national emission inventory database lacked data on CO and NH_3 emissions, which are compulsory inputs for CMAQ model, supplemental emission data on these pollutants in 2015 were obtained from the International Institute for Applied Systems Analysis (IIASA) database (at a 0.5°× 0.5° resolution) (Stohl et al., 2015). In case of the large uncertainty leaded by merging data from two datasets, the ratio of CO to VOC was checked in this study. CO and VOC emissions both result from incomplete combustion of fuel and are likely to be related (von Schneidemesser et al., 2010; Wang et al., 2014). The ratio of CO to VOC was 7.7 in the IIASA inventory and 7.5 in the final combined inventory. Thus, the CO/VOC shares in these two inventories were very close and the use of the final combined inventory is acceptable."

21. Page 8, line 29-30: "National and local emission data were allocated to simulation grids by spatial interpolation in ArcGIS 10.2 (ESRI, 2013)."

22. Page 9, line 6-8: "The initial and boundary conditions for meteorology were generated from the National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) (NCEP, 2000)"

23. Page 10, line 10-15: "However, there were also uncertainties associated with the measurements themselves and the comparison of grid-based predictions to measurements at point locations. The daily variability of simulated and observed SO_2 and $PM_{2.5}$ concentrations in four representative cities (two coastal cities and two inland cites) was displayed in Fig. S3, which indicates that the temporal variability of the simulated data was consistent with the observed data and the air quality model could capture the pollution peak in most times."

24. Page 10, line 28-29; page 11, line 1-6: "Soares et al. (2014) built a refined model for evaluating population exposures to ambient air pollution in different microenvironment. In this study, in the absence of detailed individual exposure estimates, population-weighted $PM_{2.5}$ concentrations are a better approximation of potential human exposure because they give proportionately greater weight to concentrations in areas where most people live. Population-weighted exposures have been adopted as the basis for estimating the burden of disease from air pollution in the Global Burden of Disease project run by the Institute for Health Metrics and Evaluation (Cohen et al. 2017). IHME's exposure methodology is also now used by the World Health Organization."

25. Page 11, 17-12: "We estimated 7.2 $\times10^5$ tons of annual SO_2 emissions from ships in China in 2015 taking January and June as the two reference months (see Section 2.2.1 for data description and Fig. 2a for the spatial pattern)."

26. Page 11, line 19-24: "Based on the whole year 2015 AIS data, the annual emissions of SO_2, NO_X, $PM_{2.5}$, and VOC_s from shipping sectors in YRD region were estimated at 2.2×10^5 tons (one third of the value for China), 4.7×10^5 tons, 2.7×10^4 tons, and 1.2×10^4 tons, respectively, which accounted for 7.4%, 11.7%, 1.3%, and 0.3% of the total emissions from all sources in the YRD in 2015. The emission *estimates of SO_2 and NO_X were close to Fu et al.'s estimates for 2013 year, but estimates of SO_2, NOx and $PM_{2.5}$ were slightly lower than Chen et al.'s estimates for 2014 year due to the different temporal or spatial statistical scope (Chen et al., 2019; Fu et al., 2017)."*

27. *Page 12, line 16-25: "The annual emissions of SO_2, NO_x, $PM_{2.5}$, and VOC_s from all ship-related sources within the administrative water area of Shanghai in 2015 were 4.9×10^4 tons, 1.4×10^5 tons, 6.5×10^3 tons, and 4.7×10^3 tons, respectively. The breakdown of emissions from different-type ship-related sources in Shanghai are shown in Table 3. The emissions of SO_2, NO_X, $PM_{2.5}$, and VOC_s from inland-water ships and coastal ships accounted for the majority of primary emissions from all shipping related sources in Shanghai port, ranging from 72% for VOCs to about 99% for SO_2. They comprised about 17.4% of SO_2, 24.5% of NO_x, 5.2% of $PM_{2.5}$ and 0.6% of VOC_s emissions from all pollution sources in Shanghai. The shipping emissions in Shanghai port were estimated to account for 23% of SO_2, 26% of NO_x, 23% of $PM_{2.5}$, and 28% of VOC_s from total shipping emissions in YRD. "*

28. *Page 12, liine 26-29; page 13, line 1-3: "Emissions estimates from this study fall within the range of estimates from other studies (Fu et al., 2012; Fu et al., 2017). On the basis of shipping visa data, Fu et al. (2012) determined that the total amounts of SO_2, NO_X, and $PM_{2.5}$ in the vicinity of Shanghai port in 2010 were 3.5×10^4 ton/yr, 4.7×10^4 ton/yr, and 3.7×10^3 ton/yr, respectively, substantially lower than estimates in this study. Using AIS data, Fu et al. (2017) reported 5×10^4 tons of SO_2 and 7×10^4 tons of NOx from shipping in Shanghai port in 2013, close or a bit lower than the results in this study."*

29. *Page 14, line 19-22: "The average and peak contributions from the shipping emissions in specific offshore coastal areas to the ambient SO_2 and $PM_{2.5}$ concentrations on shore for the two months are listed in Table S4. Shipping emissions beyond 12 NM had a much smaller impact on ambient SO_2, which average contributions were below 0.01 $\mu g/m^3$ and peak contributions were below 0.06 $\mu g/m^3$ (Table S4)."*

30. *Page 15, line 8-21: "Shipping emissions beyond 12 NM had limited contribution*

to SO_2 concentrations in 16 core YRD cities, implying that the boundary of 12 NM might be suitable for regulating SO_2 emissions. This could also be proved by Schembari et al., (2012), who reported that statistically significant reductions of SO_2 levels (66% to 75%) were found in 3 out of the 4 European harbours, 5 months after the implementation of the EU directive 2005/33/EC that requires all ships at berth or anchorage in European harbours use fuels with a sulfur content of less than 0.1% from January 2010. The quicker chemical reaction and shorter lifetime of SO_2 may explain why ships further out than 12 NM had much smaller impact on land ambient SO_2 concentrations (Collins et al., 2009; Krotkov et al., 2016). SO_2 reacts under tropospheric conditions via both gas-phase processes (with OH) and aqueous-phase processes (with O_3 or H_2O_2) to form sulfate aerosols, and is also removed physically via dry and wet deposition (Seinfeld and Pandis, 2006). The sulfur deposition due to shipping emissions is mainly contributed by the dry depositions (Chen et al.,2019). In the Planet boundary layer (PBL), SO_2 has short lifetimes (less than 1 day during the warm season) and are concentrated near their emission sources (Krotkov et al., 2016)."

31. *Page 16, line 4-9: "The results of these YRD analyses suggest that although ambient ship-related SO_2 concentrations were mainly affected by shipping inland or within 12 NM, expanding China's current DECA to around 100 NM or more would reduce the majority of the impacts of shipping on regional $PM_{2.5}$ pollution. It also implies that the future ECA policy should consider multiple air pollutants including the primary and secondary pollutants synchronically."*

32. *Page 17, line 12-16: "Thus, population-weighted $PM_{2.5}$ concentrations from shipping sources accounted for 0.9% to 15.5% of the population-weighted $PM_{2.5}$ concentrations from all pollution sources in June, larger than the contributions of 0.2% to 1.6% in January, which was attribute to higher shipping-related population-weighted $PM_{2.5}$ concentrations in June and higher population-weighted $PM_{2.5}$ concentrations from all pollution sources in January."*

33. *Page 19, line 11-13: "The results of the analyses of different-type shipping-related sources indicated that ship-related sources close to densely-populated areas*

contribute substantially to population exposures to air pollution."

34. *Page 19, line 14-29; page 20, line 1-11:*

*"**3.4 Limitations and uncertainties***

Limitations in the study were mainly related to some missing information in database and assumptions during estimation of shipping emissions. When estimating shipping emission inventory, underestimations of actual emissions may be introduced by missing information. For example, AIS data has a high coverage of coastal vessels, but many inland vessels are not equipped with AIS. Emissions from those inland vessels without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA, and that could introduce some uncertainties for inland river vessels. Also, emissions from fishing boats were probably underestimated because AIS devices on some fishing boats may not be in use. Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions.

In addition, we did not consider the external effects of water flow, wind, and waves when calculating engine power for ships going over the region. This would introduce some uncertainties (Aulinger et al., 2016). According to the previous studies in other areas, these factors may increase fuel consumption of individual vessels by as much as 10% to 20%, while the effects of waves on emissions estimations over extensive geographical regions are negligible (Jalkanen et al., 2009; Jalkanen et al., 2012). The downstream of the Yangtze River is located in the geographically plateau region, and the river flow is below 0.5 m/s (Song and Tian, 1997; Xue et al., 2004). For Shanghai, located at the end of mouth of the Yangtze River to the East China Sea with a flat terrain, the river flow is very slow. Given that ships traveling the Yangtze River near Shanghai have speeds over ground (SOG) of about 5-10 knots (3-5 m/s), the relative ratios of water flow to the SOG is within 20%. In our future work, we will fill the gap in the basic ship data and consider the external effects when building the shipping emission inventory.

Finally, this work only extends from emissions to air quality and population exposures. The health impacts of shipping-related air pollution in Shanghai and the YRD region will be explored in future work."

35. Page 20, line 28-29; page 21, line 1-4: "For example, policymakers could consider whether to expand China's current DECA boundary of 12 NM to around 100 NM or more to reduce the majority of shipping impacts on air pollution concentrations and exposure. It will be helpful to improve the local air quality and reduce human exposures in densely populated areas by developing more stringent regulations on the fuel quality for ships entering inland rivers or other waterways close to residential regions."

36. Page 21, line 28-30: "Aulinger, A., Matthias, V., Zeretzke, M., Bieser, J., Quante, M., and Backes, A.: The impact of shipping emissions on air pollution in the greater North Sea region - Part 1: Current emissions and concentrations, Atmos. Chem. Phys., 16, 739-758, 10.5194/acp-16-739-2016, 2016."

37. Page 22, line 22-25: "Cohen, A. J., Brauer, M., Burnett, R., Anderson, H. R., Frostad, J., Estep, K., Balakrishnan, K., Brunekreef, B., Dandona, L., and Dandona, R.: Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015, Lancet, 389, 1907-1918, 2017."

38. Page 22, line 26-27: "Collins, B., Sanderson, M. G., and Johnson, C. E.: Impact of increasing ship emissions on air quality and deposition over Europe by 2030, Meteorol. Z., 18, 25-39, 2009."

39. Page 23, line 5: "ESRI, ArcGIS 10.2. Environmental Systems Research Institute Inc, Redlands, 2013."

40. Page 23, line 13-14: "Fu, M., Liu, H., Jin, X., and He, K.: National- to port-level inventories of shipping emissions in China, Environ. Res. Lett., 12, 114024, 10.1088/1748-9326/aa897a, 2017."

41. Page 23, line 23-24: "Jalkanen, J. P. et al. Extension of an assessment model of ship traffic exhaust emissions for particulate matter and carbon monoxide. Atmos. Chem. Phys. 12, 2641–2659, 2012."

42. Page 23, line 28-30: "Krotkov, N. A., Mclinden, C. A., Li, C., Lamsal, L. N., and Streets, D. G.: Aura OMI observations of regional SO2 and NO2 pollution changes from 2005 to 2015, Atmos. Chem. Phys., 16, 4605-4629, 2016."

43. Page 24, line 1-4: "Li, C., Borken-Kleefeld, J., Zheng, J., Yuan, Z., Ou, J., Li, Y.,Wang, Y., and Xu, Y.: Decadal evolution of ship emissions in China from 2004 to 2013 by using an integrated AIS-based approach and projection to 2040, Atmos. Chem. Phys., 18, 6075–6093, https://doi.org/10.5194/acp-18-6075-2018, 2018."

44. Page 24, line 23: "Lloyd's register (IHS Fairplay). 2015."

45. Page 24, line 27-30: "Mao, X., Cui, H., Roy, B., Olmer, N., Rutherford, D., & Comer, B: Distribution of air pollution from oceangoing vessels in the Greater Pearl River Delta, 2015, The International Council on Clean Transportation, 2017. Retrieved from http://www.theicct.org/sites/default/files/publications/China-GPRD-Baseline-Emissions-Inventory_ICCT-Working%20Paper_23082017_vF.pdf"

45. Page 25, line 2-5: "Mao, X., and Rutherford, D.: NOx emissions from merchant vessels in coastal China: 2015 and 2030, The International Council on Clean Transportation, 2018. Retrieved from https://www.theicct.org/sites/default/files/publications/Merchant_Vessel_Emissions_China_20181229.pdf"

46. Page 25, line 6-9: "NCEP (National Centers for Environmental Prediction): NCEP FNL Operational Model Global Tropospheric Analyses, continuing from July 1999, Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory, https://doi.org/10.5065/d6m043c6, 2000, accessed 27 January 2018."

47. Page 25, line 21-23: "Schembari, C., Cavalli, F., Cuccia, E., Hjorth, J., Calzolai, G., Pérez, N., Pey, J., Prati, P., and Raes, F.: Impact of a European directive on ship emissions on air quality in Mediterranean harbours, Atmos. Environ., 61, 661-669, 2012."

48. Page 25, line 24-25: "Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, 2nd Edn. 2006, John Wiley &

<u>Sons, Hoboken, New Jersey, 2006.</u>"

49. Page 25, line 26-29: "<u>Soares, J., Kousa, A., Kukkonen, J., Matilainen, L., Kangas, L., Kauhaniemi, M., Riikonen, K., Jalkanen, J. P., Rasila, T., Hänninen, O., Koskentalo, T., Aarnio, M., Hendriks, C., and Karppinen, A.: Refinement of a model for evaluating the population exposure in an urban area, Geosci. Model Dev., 7, 1855-1872, 10.5194/gmd-7-1855-2014, 2014</u>"

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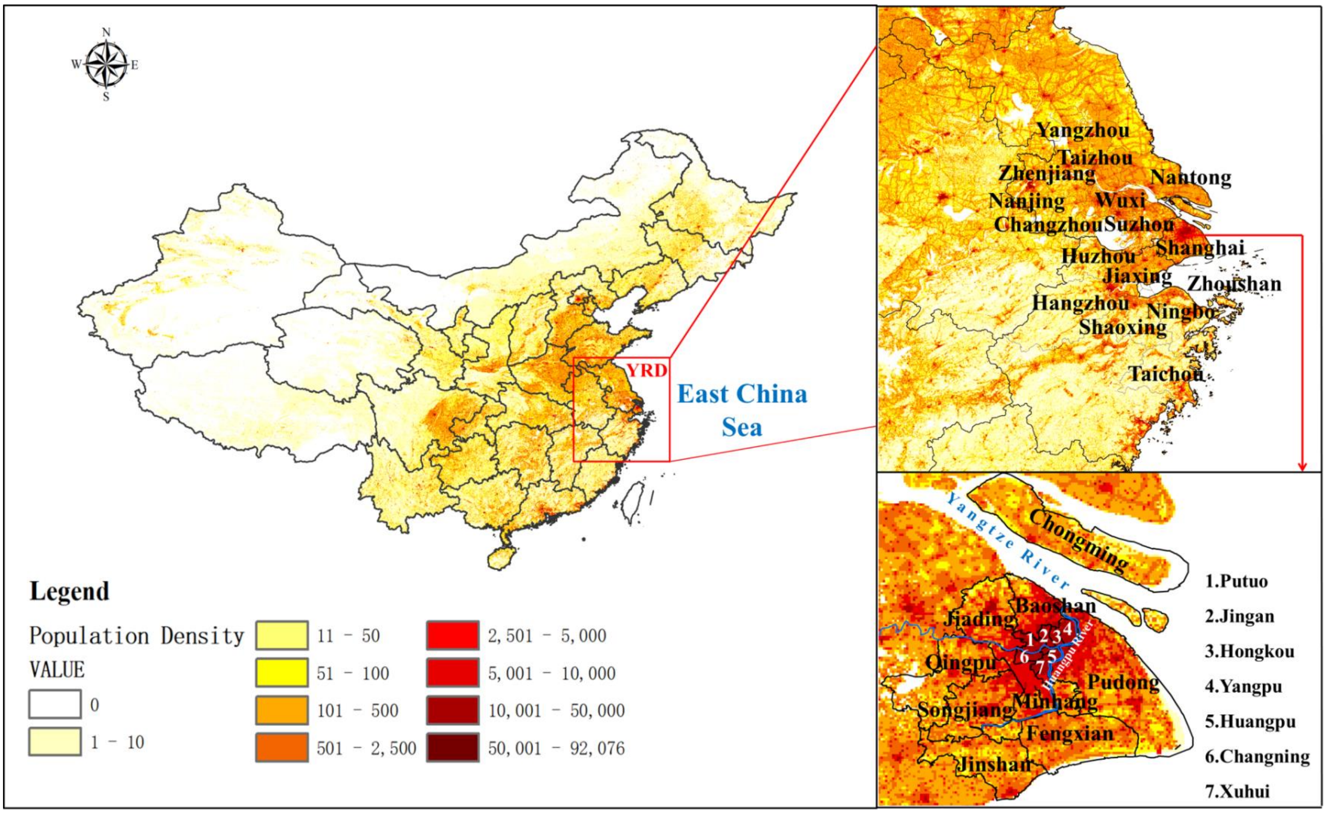
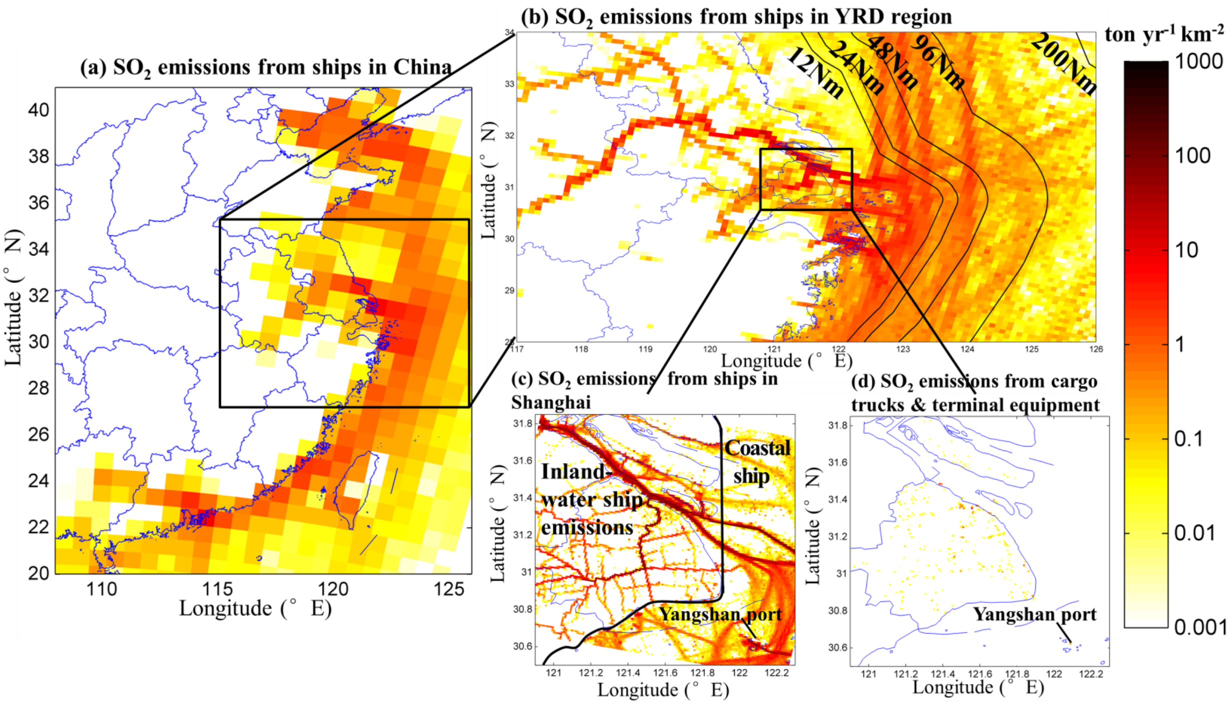
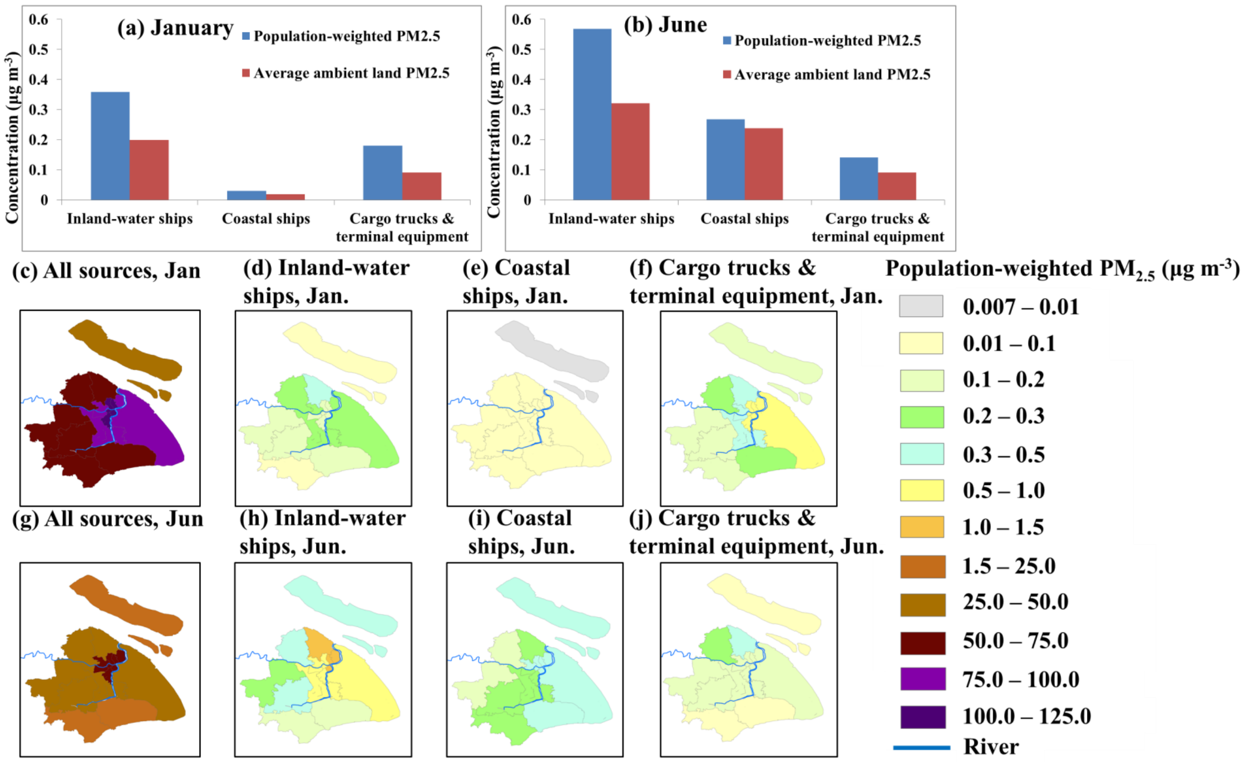
55. Figure 1. Geographic location of the study area YRD/Shanghai with population density in 2015. 16 core cities in YRD and 16 administrative districts in Shanghai are noted on the map. The smaller administrative districts are labeled with numbers: Putuo (1), Jingan (2), Hongkou (3), Yangpu (4), Huangpu (5), Changning (6), Xuhui (7).



56. Figure 2. SO_2 emissions in 2015 from (a) shipping traffic in China (the average value of January and June) at resolution of 81km × 81km; (b) ships in different offshore coastal areas (inland-water and within 12 NM, 12-24 NM, 24-48 NM, 48-96 NM and 96-200 NM) in the YRD region, at resolution of 9km × 9km; (c) inland-water ships and coastal ships in Shanghai, at resolution of 1km × 1km; and (d) container-cargo trucks and port terminal equipment in Shanghai, at resolution of 1km × 1km. The black line in (c) refers to the division line between the inland water and coastal water for Megacity Shanghai defined in this study.



57. Figure 8. Population-weighted $PM_{2.5}$ and average $PM_{2.5}$ caused by different ship-related sources in Shanghai, in January(a) and in June (b); population-weighted $PM_{2.5}$ caused by all pollution sources (c, g), inland-water ships (d, h), coastal ships (e, i) and container-cargo trucks and port terminal equipment (f, j) in 16 districts in Shanghai, in January 2015(c-f) and June 2015 (g-j).



58. Table 1 Statistical metrics of the model evaluation. Observed data (Obs.) and simulated data (Sim.) for each city are the average of monthly values of January and June case. NMB, NME, RMSE and r were calculated based on the daily-average observed and simulated data.

59. Table 2, caption: Primary emissions (ton/yr), emission share in all shipping emissions (%) and emissions density (ton/yr/km^2) from shipping at different boundaries in YRD region[a] in 2015

60. Table 3, caption: Primary emissions (ton/yr) and emission share in all pollution sources (%) from different-type ship-related source in Shanghai[a] in 2015

61. Supporting information, page 1, line 30-31: "S2 Fuel type, sulfur content, engine type, emission Factors, low load adjustment multipliers, and control factors"

62. Supporting information, page 2, line 28-30: "However, auxiliary boiler emissions were not considered in this study because limited auxiliary boiler information exists in the Lloyd's register and Chinese Classification Society (CCS) database."

63. Supporting information, page 2, line 35-37; page 3, line 1-7: "For ships available in Lloyd's register (now IHS-Fairplay) (Lloyds, 2015) and CCS database, the following data were derived from these database including: ship name, ship type, date of construction, flag name, revolutions per minute (RPM) of the main engine, speed, maximum design power of the main engine, maximum design power of the auxiliary engines and gross tonnage. Information of some domestic ships is available in CCS database, but for those ships unavailable in the database, the main engine power was assumed to be 7000 kw by default, which was close to the domestic ships from the CCS database (with main engine power mainly ranging from 4000 kw to 6000 kw) and below the East China Sea-going ships in Lloyd's register (with main engine power mainly ranging from 11000 kw to 14000 kw)"

64. Supporting information, page 3, line 11-27:

*"**S.2 Fuel type, sulfur content, engine type, emission Factors, low load adjustment multipliers, and control factors***

The two most common fuel oils used in ships are residual oil (RO) and marine distillates (MD). In general, RO is used in the main engine, and the fuel sulfur content is approximately 2.7%, MD is used in the auxiliary engine, and the sulfur content is approximately 0.5%. On the basis of data on ships passing by the Port of Shanghai provided by the largest Chinese heavy fuel oil (HFO) supplier, China Marine Bunker (CMB), the sulfur content of the fuel used by the main engines in domestic vessels *ranges from 0.2% to 2.0%, and the sulfur content of the fuel used by the main engines in ocean-going vessels ranges from 1.9% to 3.5%. In this study, we adjusted the sulfur content of the fuel used by the main engines in domestic vessels to 1.5% and that of ocean-going vessels to 2.7%. The amount of SO_2 emitted is directly affected by the sulfur content of the fuel; therefore, when main engine emissions were estimated by the model, the emissions of domestic vessels were amended correspondingly. The main engine category was sorted into slow speed diesel (SSD), medium speed diesel (MSD), and high speed diesel (HSD) based on the engine revolutions per minute (RPM), and the largest auxiliary engine category was MSD. The type of engine was judged first according to the RPM of the main engine in Lloyd's database. The emission factors of the different types of engines differ considerably."*

65. Supporting information, page 3, line 33-34: "Emission factors used in the present study were listed in Fan et al. (2016)."

66. Supporting information, page 3, line 36-39; page 4, line 1-2: "Because adjustment multipliers were not available for organic carbon (OC) and elemental carbon (EC), these pollutants were assigned the same low load adjustment multiplier (LLAM) as PM in the present study, which may introduce uncertainties. In this study, the ratio of BC emissions to PM emissions (BC/PM) was around 2.9%, which falls within the range of 2% to 4.5% in other studies (Comer et al., 2017; Erying et al., 2005; Petzold et al., 2004)."

67. Supporting information, page 5, line 7-11: "Comer, B., Olmer, N., Mao, X., Roy, B., and Rutherford, D.: Black Carbon Emissions and Fuel Use in Global Shipping, 2015, The International Council on Clean Transportation, 2017. Retrieved from https://www.theicct.org/sites/default/files/publications/Global-Marine-BC-Inventory-2015_ICCT-Report_15122017_vF.pdf"

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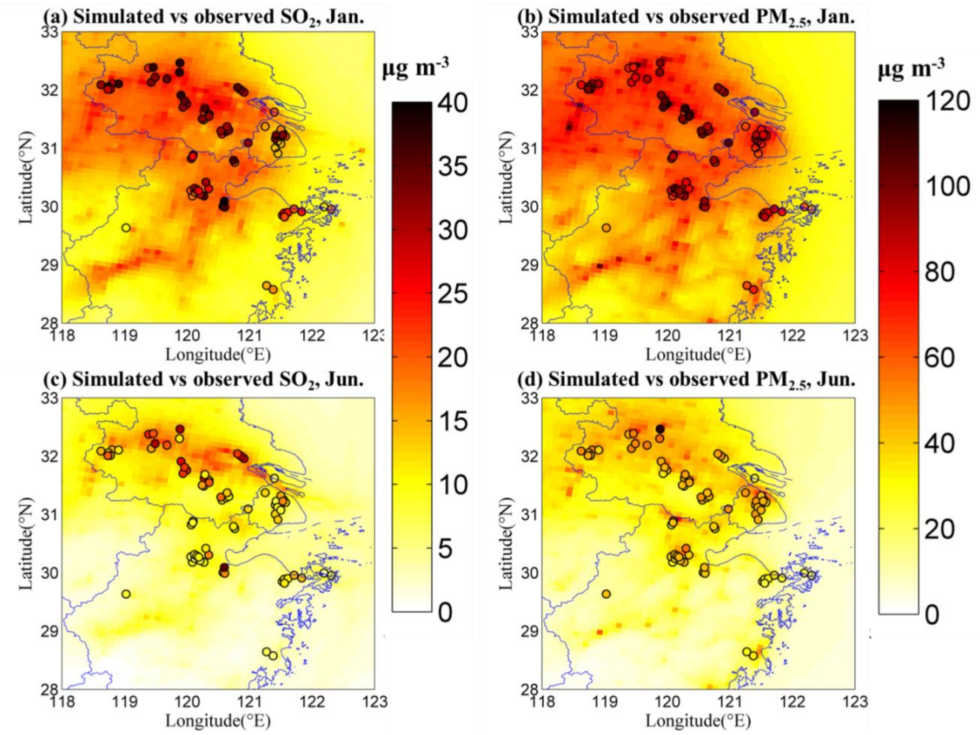
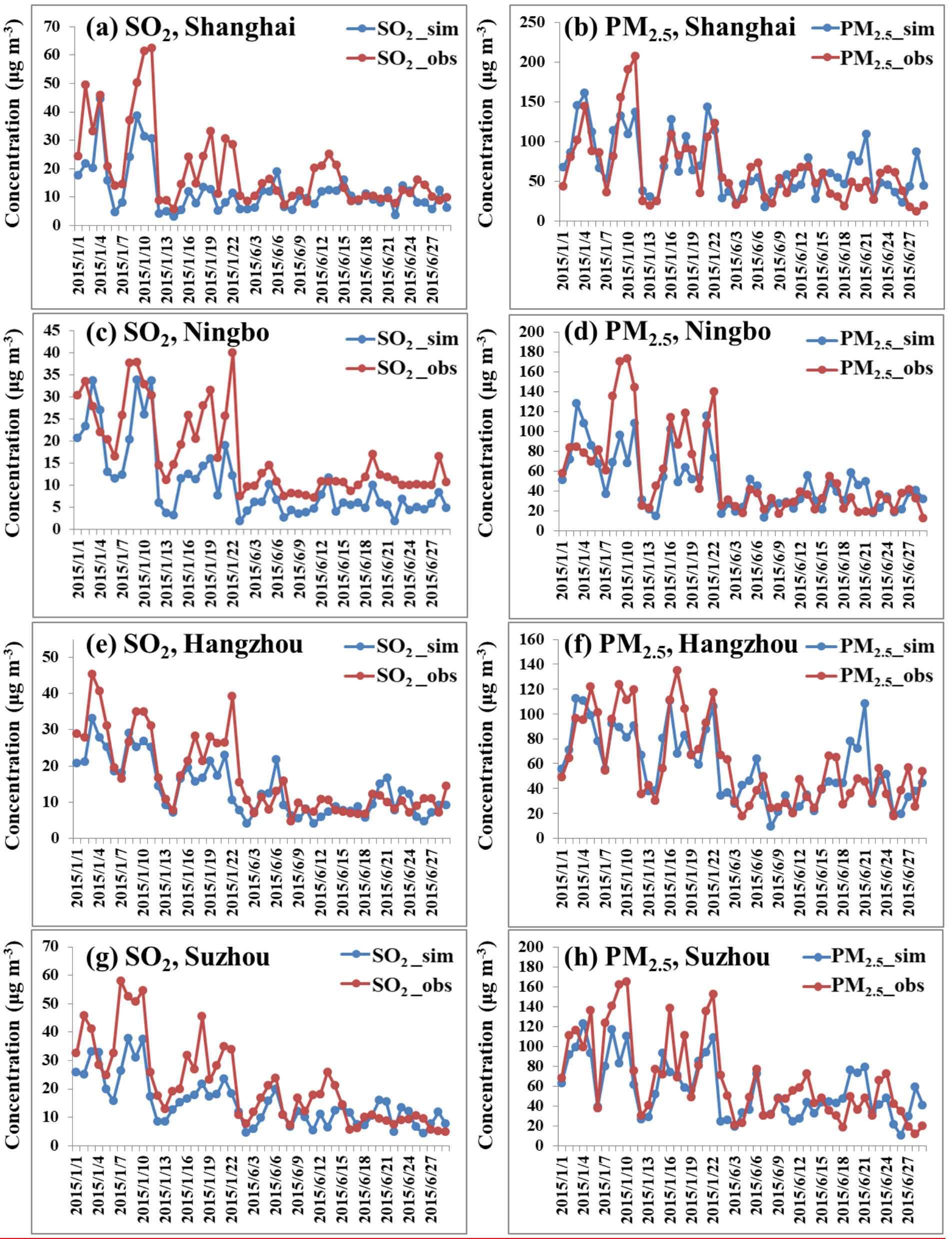
73. Supporting information: Figure S2. The simulated (grid) and observed (circles) SO_2 concentration distribution in YRD region, in January 2015 (a) and June 2015 (c); the simulated (grid) and observed (circles) $PM_{2.5}$ concentration distribution in YRD region, in January 2015 (b) and June 2015 (d)



74. Supporting information: Figure S3 Daily variability of simulated (sim.) and observed (obs.) SO_2 concentrations (a, c, e, g) and $PM_{2.5}$ concentrations (b, d, f, h) in four representative cities, including two coastal cities – Shanghai (a, b) and Ningbo (c, d), and two inland cites – Hangzhou (e, f) and Suzhou (g, h).



75. Supporting information: Table S4 Average and peak contributions from ship emissions in different offshore coastal areas to the ambient SO_2 and $PM_{2.5}$ concentrations in January and June

Offshore distance	*Average contribution (μg/m^3^)*				*Maximum contribution (μg/m^3^)*			
	SO_2		$PM_{2.5}$		SO_2		$PM_{2.5}$	
	January	*June*	*January*	*June*	*January*	*June*	*January*	*June*
Inland and within 12 NM	*0.52*	*0.70*	*0.24*	*0.56*	*6.00*	*8.79*	*1.62*	*4.02*
12-24 NM	*0.005*	*0.007*	*0.01*	*0.04*	*0.03*	*0.05*	*0.05*	*0.20*
24-48 NM	*0.01*	*0.009*	*0.04*	*0.07*	*0.06*	*0.05*	*0.11*	*0.34*
48-96 NM	*0.02*	*0.008*	*0.07*	*0.07*	*0.05*	*0.03*	*0.14*	*0.30*
96-200 NM	*0.00*	*0.001*	*0.003*	*0.01*	*0.004*	*0.003*	*0.02*	*0.05*

The influence of spatiality on shipping emissions, air quality and potential human exposure in Yangtze River Delta/Shanghai, China

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Abstract: The Yangtze River Delta (YRD) and the megacity of Shanghai are host to one of the busiest port clusters in the world; the region also suffers from high levels of air pollution. The goal of this study was to estimate the contributions of shipping to regional emissions, air quality, and population exposure and to characterize the importance of the geographic spatiality of shipping lanes and different-type shipping-related sources ~~ir dependence on the geographic spatiality of ship lanes from the regional scale to city scale~~ for the baseline year 2015, prior to the implementation of China's Domestic Emission Control Areas (DECAs) in 2016. The WRF-CMAQ model was used to simulate the influence of coastal and inland-water shipping, in port emissions, shipping-related cargo transport on air quality and, population-weighted concentrations, a measure of human exposure. Our results showed that the impact of shipping on air quality in the YRD was attributable primarily to shipping emissions within 12 NM of shore, but emissions coming from the coastal area of 24 to 96 NM still contributed substantially to ship-related $PM_{2.5}$ concentrations in YRD. The overall contribution of ships to $PM_{2.5}$

concentration in YRD could reach to 4.62 μg/m^3 in summer when monsoon winds transport shipping emissions onshore. In Shanghai city, inland-water going ships were major contributors (40-80%) to the shipping impact on urban air quality. Given the proximity of inland-water ships to urban populations of Shanghai, the emissions of inland-water ships contributed more to population-weighted concentrations. These research results provide scientific evidence to inform policies for controlling future shipping emissions; in particular, in the YRD region, expanding the boundary of 12 NM in China's current DECA policy to around 100 NM would include most of the shipping emissions affecting air pollutant exposure, and stricter fuel standards could be considered for the ships on inland rivers and other waterways close to residential regions.

Key words: Shipping, ports, emissions, source apportionmentattribution, population-weighted concentration, Shanghai, Yangtze River Delta /YRD, emission control area

1 Introduction

With the increase of international maritime trade, shipping emissions and their impacts have attracted increased attention globally over the past decades (Capaldo et al., 1999;Cooper, 2003;Eyring et al., 2010;Sofiev et al., 2018). Shipping emits air pollutants that contribute to adverse impacts on climate, on air quality and on the health of people living near ports (Li et al., 2018;Liu et al., 2016a). Globally, about 50,000 to 90,000 cardiopulmonary diseases and lung cancer deaths due to cardiopulmonary diseases and lung cancer each year are were attributable to exposure to particulate matter emitted from shipping in 2010 and 2012, respectively (Corbett et al., 2007;Partanen et al., 2013;Winebrake et al., 2009),. and 403,300 premature mortalities per year due to shipping are predicted in 2020 under business-as-usual (BAU) assumptions (Sofiev et al., 2018). In Europe, ozone pollution caused by international ships led to around 3.6 % of the total estimated years of life lost and 2.6 % of premature deaths in 2005 (Campling et al., 2013). In East Asia, around 14,500 to 37,500 premature deaths per year has been primarily attributed to $PM_{2.5}$ from shipping; about one third of those deaths were in the area surrounding the East China sea, with the largest impacts in mainland China (Liu et al., 2016a).

As of 2016, China was home to 7 of the top 10 container ports, and the size of those ports has been rapidly growing to serve the increased trade via international shipping (UNCTAD, 2017). The Yangtze River Delta (YRD) is one of the economic centers as well as home to the busiest port clusters, comprised of more than 15 ports, including Shanghai port, Ningbo-Zhoushan port, Zhenjiang port, Nantong port, Lianyungang port, Taizhou port, and Wenzhou port. In 2016, YRD generated a GDP of RMB 17.72 trillion (US $2.76 trillion) – about 20 percent of China’s national GDP (Preen, 2018). Shanghai megacity itself is an important economic center, accounting for about 22 % of the total GDP in YRD. Shanghai port lies at the intersection of the East China Sea and the Yangtze River and has been the largest container port in the world since 2010 (Liu et al., 2016b).

Shanghai and the YRD are also among the most densely populated regions of China. The YRD is home to 239.1 million people; Shanghai is one of the largest cities and houses about 12.1 % of the total population of the YRD (Bright et al., 2016).

This region has suffered from severe air pollution over the past decade due to the anthropogenic emissions from multiple sources. In December 2013, for example, YRD experienced a haze episode, in which the maximal observed $PM_{2.5}$ concentration in YRD exceeded 590 $\mu g/m^3$ (Sun et al., 2016). As severe air pollution episodes have continued and ports have grown, the shipping sector, a subset of transportation pollution sources, has received more attention.

The high ship traffic density in Shanghai and YRD has led to high emissions of shipping-related air pollutants in this region (Fan et al., 2016). Shipping-related sources of air pollution in Shanghai comprise coastal ships, inland-water ships, container-cargo trucks, and port terminal equipment. Because some of these emissions sources are also close to densely populated areas, in particular those from ships traveling in inland waterways and from container trucks transporting cargo in and around the city, there is greater potential for higher population exposures to ship-related air pollution.

The International Maritime Organization (IMO) regulates emissions of marine pollution on a global scale. Current rules limit fuel sulfur content (FSC) to 3.5 % globally and will lower this limit to 0.5 % in 2020. The IMO has also designated several regional Emission Control Areas (ECAs) to benefit the atmospheric environment and human health in port and coastal communities that establish more stringent emissions limits up to 200 NM from the coast in the Baltic Sea (SO_X), North Sea (SO_X), North America (SO_X, NO_X, and PM), and the United States Caribbean Sea area (SO_X, NO_X, and PM) (Viana et al., 2015). Fuel sulfur content is limited to 0.1 % in the ECAs.

China does not have an ECA designated by the IMO, but in December 2015 it designated three Domestic Emission Control Areas (DECAs) that operate in a similar manner. These DECAs limited fuel sulfur content to 0.5 % for ocean-going vessels (OGV) in 3 regions: YRD, Pearl River Delta (PRD) and Bohai Sea. The DECA implementation timeline specified that qualified ports are encouraged to be in compliance since April 1, 2016, and all ships at berth in 11 core ports within these regions would be in compliance by January 1, 2017 and all ocean-going vessels (OGV) or coastal vessels within 12 NM of the shoreline would be in compliance by January 1, 2019. These areas would also be in compliance with the IMO requirements for fuel sulfur content. A study reported that the average reduction of $PM_{2.5}$ and SO_2 mass concentrations over land in the PRD due to the DECA policy were 2.7% and 9.54% (Liu et al., 2018a). China is currently considering additional DECA restrictions for the period beyond 2019. Starting on October 1, 2018, three months earlier than the original plan, the Shanghai Maritime Safety Administration (MSA) has enforced the DECA policy limiting fuel sulfur content to 0.5 % for ocean-going vessels and domestic coastal vessels in Shanghai port. However, the DECA policies for fuel sulfur content currently make no distinction between coastal ships that enter inland water areas and other ships. Ships like those in Shanghai and the YRD that enter inland waterways bring emissions sources closer to population centers resulting in a greater potential for exposure and health impacts.

Shipping emission inventories for the YRD, PRD, and Bohai-Rim area and their major ports indicate that shipping is an important pollution source surrounding port regions (Chen et al., 2016;Fan et al., 2016;Li et al., 2016;Yau et al., 2012). Several studies have investigated the contribution of shipping emissions to ambient air quality using different methods. Zhao et al. (2013) analyzed aerosol samples in Shanghai Port and reported that ship traffic contributed 0.63 μg/m^3 to 3.58 μg/m^3 (or 4.2 % to 12.8 %) of the total $PM_{2.5}$ in Shanghai Port. ~~Using half-hour aerosol time-of-flight mass spectrometer measurements, Liu et al. (2016b) estimated that the number concentrations of primary~~ Primary ship-emitted particles measured by an aerosol time-of-flight mass spectrometer were typically ~~contributed~~ 1.0 to 10.0 % of the measured particle number concentration, with the contribution rising to as high as 50.0 % in spring and summer (Liu et al. 2016b). ~~Tao et al. (2016) reported that shipping emissions were among the top contributors to $PM_{2.5}$ i~~In Guangzhou and Zhuhai, shipping emissions were among the top contributors to $PM_{2.5}$ and accounted~~ing~~ for greater than 17% of $PM_{2.5}$ mass concentrations (Tao et al. 2016). Using

WRF-CMAQ, Chen et al. (2017b) estimated found that the contribution of shipping emissions to the $PM_{2.5}$ mass concentrations in Qingdao is was the highest in summer (13.1%) and the lowest in winter (1.5 %). Chen et al. (2019) reported that ship traffic sources could contribute 4.0 % of annual $PM_{2.5}$ mass concentrations over the land area in YRD and the maximum could reach 35.0% in port region in 2014.

In China, a few studies reported the contribution to air pollution from shipping in different offshore coastal areas or different-ta ype ship-related sources. For example, Mao et al. (2017) estimated primary emissions from OGVs at different boundaries in the PRD region, and concluded that further expansion of emission control area to 100 NM would provide even greater benefits. However, the impacts of shipping emissions at varying distances from shore on air quality and potential human exposure, which are important when considering ECA policy, have not been rigorously studied. Mao and Rutherford (2018) studied NO_x emissions from three categories of merchant vessels—OGVs, coastal vessels (CVs) and river vessels (RVs) in China's coastal region. But less attention was paid to the impacts of inland waterway traffic and port-related sources like container-cargo trucks and terminal port equipment on air quality and potential human exposure.

To fill this gap, Tthe overall goal of this study was to characterize the spatial distribution of shipping-related emissions and their impacts on air quality and human exposure in the YRD and Shanghai for the baseline year 2015, prior to the implementation of China's DECAs in 2016. We modeled shipping emissions in different offshore areas in the YRD region and emissions from different-type ship-related sources in Shanghai city for each month of the year. For YRD region, we focused on shipping emissions in different offshore areas, while for Shanghai city, we studied individual ship-related sources in areas under the jurisdiction of Shanghai MSA. To identify which offshore areas in the YRD region and which ship-related sources in Shanghai contributed the most ambient air pollution, and human population exposure, we modeled the impacts of shipping emissions in different offshore areas (within 12 NM including inland waters, 12-24 NM, 24-48 NM, 48-96 NM, and 96-200 NM) in the YRD region as well as coastal ships, inland-water ships, and container-cargo trucks and port terminal equipment in and near the port areas under the jurisdiction of Shanghai MSA in two representative months (January and June). The impacts were evaluated for two different months, January and June, to represent seasonal differences. The results of this study could be informative to the consideration of the distance of regulated emissions in the design of future emissions control areas for shipping in YRD, or regulations on the sulfur content of fuels for different-type ship-related sources in Shanghai.and its related sources in Shanghai/YRD.

2 Methodology

In this study, we first established a shipping emission inventory based on highly-resolved automatic identification system (AIS) data in 2015. Then, we used WRF-CMAQ model to evaluate the impacts on air quality from shipping emissions in different offshore coastal areas (within 12 NM including inland waters, 12-24 NM, 24-48 NM, 48-96 NM, and 96-200 NM) in the YRD region. We referred to ICCT's working paper (Mao et al., 2017) to choose the distance bins between 12 NM (the boundary of current China's DECA) and 200 NM (the boundary of ECA designated by IMO) in this study. The model domains were shown in Figure S1. Simulations were also conducted to estimate the influence of individual different-type shipping-related sources (coastal ships, inland-water ships, container-cargo trucks, and port terminal equipment) on air quality in Shanghai. Finally, population-weighted $PM_{2.5}$ concentrations attributable to shipping sources were calculated.

2.1 Study area and period

Figure 1 shows the geographic area and population density for the YRD and Shanghai, the location of 16 core cities of the YRD region, and 16 administrative districts within Shanghai city. The coastal cities in the YRD are Nantong, Shanghai, Jiaxing, Ningbo, Taichou, and Zhoushan.

The simulation network was developed for four domains at resolutions of 81 km × 81 km, 27 km × 27 km, 9 km × 9 km, and 1 km × 1 km, respectively (Fig. S1). Domain 1 covers East Asia and part of south east Asia the whole China. Nested domains 2, 3, and 4 cover a large part of East-China (2), the YRD region (3, including Jiangsu, Zhejiang, and Shanghai), and Shanghai with a finer resolution (4), respectively. The geographic scope for the YRD study area extended from 116.5 °E to 127 °E and 27 °N to 35 °N and included an offshore distance of approximately 200 NM. The Shanghai study area included from 120.5 °E to 122.3 °E and from 30.5 °N to 32 °N, where the water is within the jurisdiction of Shanghai MSA (up to about 12 NM from shore).

Two contrasting representative months in the year 2015, January and June, were selected to compare the seasonal effects. The highest shipping impacts were expected in June because shipping activity and emissions are higher in summer than at other times of year (Fan et al., 2016;Jalkanen et al., 2009) and prevailing winds from the summer monsoon are directed from the ocean to the shore. Higher shipping impacts were expected in June because prevailing winds from the summer monsoon are directed from the ocean to the shore. January was chosen as a contrasting period with prevailing winds away from shore.

2.2 Shipping eEmission inventories

2.2.1 Ship-related emission inventories

In this study, emission inventories were constructed based primarily on automatic identification system (AIS) data for all shipping traffic activity in China, YRD, the Shanghai and Shanghai YRD geographic domains. Due to limitation of the national-scale data source, the AIS data in this study only covered the representative months of January and June 2015, while the YRD-scale AIS data covered 2015 full year. AIS data includes international ships, coastal ships, and inland-water ships, but some river ships could be not covered by AIS data.— Emissions from ships entering the geographic domains for YRD or Shanghai were calculated using the AIS-based model developed by Fan et al. (Fan et al., 2016), and monthly shipping emissions for January and June were used in the air quality model to capture the seasonal variation to expect more accurately than annual shipping emissions with no monthly variations.. For Shanghai, estimates of emissions from those ships without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA. The detailed method, assumptions and sources are provided in section S.1 of the supplemental materials supporting information. The actual speeds and operation times of the ships involved in the calculation can be obtained from AIS data with high accuracy, while the installed power of the main engine (ME), auxiliary engine (AE), and auxiliary boiler (AB) and the maximum speed of ships necessary to complete the estimates were obtained from Lloyd's register (now IHS-Fairplay) (Lloyds, 20092015) and the China Classification Society (CCS) database. Assumptions regarding the fuel types, sulfur contents and engine types, and sSources of emission factors, low load adjustment multipliers, and control factors are provided in section S.2 of the supporting information. Values of these factors can be found in our earlier study (Fan et al., 2016). We assumed that the sulfur content of the fuel burned by the main engines was 2.7% for international coastal ships, and 1.5% for domestic coastal ships.

Within the Shanghai port domain, separate emissions inventories were developed to estimate the relative air quality impacts of coastal and inland-water and of ship-related container-cargo trucks transport and port terminal equipment (cranes, forklifts, and trucks used for internal transport). Many coastal ships operate in both the outer port and in the inner river region of Shanghai Port, which includes the Yangtze River, Huangpu River and other rivers in Shanghai. Consequently, a geographic boundary was used to divide the shipping emissions inventory based on AIS data into coastal and inland sources (see Figure 3c in which the black line denotes a division between coastal and inland shipping contributions to emissions).

Emissions from container-cargo trucks were estimated using International Vehicle Emission (IVE) model (Wang et al., 2008). The vehicular activity data was provided by the Shanghai Traffic Department. The emissions from port terminal equipment including the trucks in port were calculated based on fuel consumption for each part of the port. Given their smaller emissions relative to shipping and other non-port sources, emissions from container-cargo trucks and terminal equipment were combined and gridded at a resolution of 1 km × 1 km.

2.2.2 Non-shipping emission inventories

National and local YRD emission inventories were used for emissions from all other sources (non-shipping). For the national scale domain, we used a 2015 national emission database at a 27 km × 27 km resolution that included 5 pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_x and VOCs) and 14 source types (see Table S1 for details) (~~Fu et al., 2013~~ Zhao et al., 2018). Since the national emission inventory database lacked data on CO and NH_3 emissions, which are compulsory inputs for CMAQ model, supplemental emission data on these pollutants in 2015 were obtained from the International Institute for Applied Systems Analysis (IIASA) database (at a 0.5 °× 0.5 ° resolution) (Stohl et al., 2015). In case of the large uncertainty leaded by merging data from two datasets, the ratio of CO to VOC was checked in this study. CO and VOC emissions both result from incomplete combustion of fuel and are likely to be related (von Schneidemesser et al., 2010; Wang et al., 2014). The ratio of CO to VOC was 7.7 in the IIASA inventory and 7.5 in the final combined inventory. Thus, the CO/VOC shares in these two inventories were very close and the use of the final combined inventory is acceptable. The Shanghai Academy of Environmental Sciences (SAES) provided the local YRD land-based emission inventory at a 4 km × 4 km resolution; it included 8 source types and 7 pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_x, CO, VOCs and NH_3). Details are provided in Table S2. National and local emission data were allocated to simulation grids by spatial interpolation in ArcGIS 10.2 (ESRI, 2013).

2.3 WRF-CMAQ model setup

The models used in this study were the Weather Research and Forecasting Model (WRF) version 3.3 and the Community Multiscale Air Quality (CMAQ) model version 4.6. The selected simulation periods were 1 January to 31 January and 1 June to 28 June, with 72 hours of spin-up time for each run. The initial and boundary conditions for meteorology were generated from the ~~Chinese~~ National Centers for Environmental Prediction (NCEP) Final Analysis (FNL) (NCEP, 2000) with resolution at 1 °×1 ° at six hour time intervals. Vertically, 27 sigma layers were set for the WRF simulation, and the results were then converted to the 24 layers required by CMAQ (version 4.6) using the MICP (Meteorology-Chemistry Interface Processor). CMAQ was configured to use the Carbon Bond mechanism (CB05) for gas-phase chemistry and the AERO4 aerosol module (Liu et al., 2016b).

2.4 Simulations of source contribution to air quality

Individual source contributions to gridded ambient concentrations of air pollution were estimated as the difference between the concentrations simulated with all sources included and those with the individual source excluded. For the YRD region (domain 3), the simulation was conducted for ships within different boundaries from shore (12 NM, 12-24 NM, 24-48 NM, 48-96 NM and 96-200 NM). For the city of Shanghai, simulations were conducted for all ship-related sources in the water area under the jurisdiction of Shanghai MSA (within approximately 12 NM of shore), coastal and inland-water shipping (as defined geographically above), and container-cargo transport and port terminal equipment (combined). Details of each simulation can be found in Table S3.

2.5 Model evaluation

Performance of the models was spatially evaluated by comparison with monthly-average observations at monitoring stations (Fig. S2). Generally, the simulated results showed trends consistent with the observations, with increased concentrations of SO_2 and $PM_{2.5}$ along the Yangtze River and in the urban areas. Also, daily-average observations from 53 monitoring stations in 16 core YRD cities were compared with daily-average simulated ambient SO_2 and

$PM_{2.5}$ concentrations. Normalized Mean Bias (NMB), Normalized Mean Error (NME), Root Mean-square Error (RMSE), and Pearson's correlation coefficient (*r*) were used to qualify the degree of deviation between the observed data and modeling results (Eder and Yu, 2007). Detail equations of the above statistical metrics are shown in section S.3. For each of the cities, the statistical metrics were calculated based on the average observed data and simulated results of the monitoring stations in the city, as shown in Table 1. For most cities, SO_2 and $PM_{2.5}$ concentrations were underestimated to varying degrees, which NMB was in the range of -36% to -18% and -34% to 8%, respectively. The deviations between the simulation results and the monitoring data were mainly due to the uncertainties of emission inventories and some deficiencies of meteorological and air quality models. However, there were also uncertainties associated with the measurements themselves and the comparison of grid-based predictions to measurements at point locations. The daily variability of simulated and observed SO_2 and $PM_{2.5}$ concentrations in four representative cities (two coastal cities and two inland cites) was displayed in Fig. S3, which indicates that the temporal variability of the simulated data was consistent with the observed data and the air quality model could capture the pollution peak in most times.

2.6 Population-weighted $PM_{2.5}$ concentration

~~To provide a better estimate of human exposure to shipping-related air pollution, this study estimated~~ We estimated population-weighted $PM_{2.5}$ concentrations for the 16 cities of the YRD region and the 16 districts with Shanghai city. The population-weighted $PM_{2.5}$ concentration of the given grid cell *i* is calculated based on Eq. (1) (Prasannavenkatesh et al., 2015):

$$\text{Population-weighted } PM_{2.5} \text{ concentration} = \sum_{i=1}^{n}\left(PM_i \times \frac{P_i}{\sum_{i=1}^{n} P_i}\right) \quad (1)$$

where, PM_i is defined as the $PM_{2.5}$ concentration in the *i*th grid cell, P_i is the population in the *i*th grid value of, and *n* is the number of grid cells in the selected geographical area, for example city or region. ~~Population-weighted $PM_{2.5}$ concentrations are a better approximation of potential human exposure because they give proportionately greater weight to concentrations in areas where most people live.~~

Soares et al. (2014) built a refined model for evaluating population exposures to ambient air pollution in different microenvironment. In this study, in the absence of detailed individual exposure estimates, population-weighted $PM_{2.5}$ concentrations are a better approximation of potential human exposure because they give proportionately greater weight to concentrations in areas where most people live. Population-weighted exposures have been adopted as the basis for estimating the burden of disease from air pollution in the Global Burden of Disease project run by the Institute for Health Metrics and Evaluation (Cohen et al. 2017). IHME's exposure methodology is also now used by the World Health Organization.

3 Results and Discussion

3.1 Characteristics of shipping emissions

~~Based on the average emissions from January and June, w~~We estimated 7.2 $\times 10^5$ tons of annual SO_2 emissions from ships in China in 2015 taking January and June as the two reference months (see Section 2.2.1 for data description and Fig. ~~3a~~ 2a for the spatial pattern). Below, we discuss the quantity and other characteristics of primary emissions from ships in different offshore coastal areas in YRD regions (Section 3.1.1) and from different ship-related sources in Shanghai (3.1.2).

3.1.1 Shipping emissions in YRD region

Based on the whole year 2015 AIS data, the annual emissions of SO_2, NO_X, ~~and~~ $PM_{2.5}$, and VOC_s from shipping sectors in YRD region were estimated at 2.2$\times 10^5$ tons (one third of the value for China), 4.7$\times 10^5$ tons, ~~and~~ 2.7$\times 10^4$ tons, and 1.2$\times 10^4$ tons, respectively, which accounted for 7.4%, 11.7%, ~~and~~ 1.3%, and 0.3% of the total emissions from all sources in the YRD in 2015. The emission estimates of SO_2~~,~~ and NO_X~~, and $PM_{2.5}$~~ were close to Fu et al.'s estimates for 2013 year, but estimates of SO_2, NOx and $PM_{2.5}$ were ~~a bit~~slightly lower than ~~Fan et al.'s estimates for 2010 year and~~ Chen et al.'s estimates for 2014 year due to the different temporal or spatial statistical scope (Chen et al., 2019; ~~Fan et al., 2016~~ Fu et al., 2017). However, the proportion of ship SO_2 emissions of YRD region accounting for the whole China in this study is consistent with the 33%~~-~~ to 37% in the other studies (Chen et al., 2017a; Chen et al., 2019; Liu et al., 2018b; Lv et al., 2018)

More than 60% of annual emissions of SO_2 from ships in the YRD occurred inland or within

12 NM of shore, where 75.0% of the NO_x emissions and 48.4% of the $PM_{2.5}$ emissions from ships occurred (Table 2). Similar results were obtained in a study in the PRD in 2016 other studies (Li et al., 2018; Liu et al., 2018a). Our estimate of 1.3×10^5 tons of annual SO_2 emissions emitted by ships on inland waters or within 12 NM of shore was 47% higher than Liu et al.'s estimate of 8.83×10^4 tons. However, our estimate of average emission intensity of SO_2 within 12 NM of shore in the YRD was 0.66 ton/yr/km^2, much lower than Liu et al.'s estimate of 4.04 ton/yr/km^2. One explanation for the different results may be that the YRD has a longer coastline than the PRD which leads to larger total emissions but to lower intensity. Emissions occurring within 24-48 NM and 48-96 NM from shore were not negligible; annual SO_2 emissions in these two areas accounted for 11.4% and 14.9% of the total shipping emissions in the YRD, respectively. The spatial pattern of annual SO_2 emissions from ships varied in different offshore coastal areas in the YRD (Fig. 3b2b). SO_2 emissions were also high at the intersection of Yangtze River and Huangpu River, between 24 and 48 NM from shore and in the north-south shipping lanes between 48 and 96 NM from shore.

3.1.2 Emissions from different ship-related sources in Shanghai

The annual emissions of SO_2, NO_x, and $PM_{2.5}$, and VOC_s from all ship-related sources within the administrative water area of Shanghai in 2015 were 4.9×10^4 tons, 1.4×10^5 tons, and 6.5×10^3 tons, and 4.7×10^3 tons, respectively. The breakdown of emissions from individual different-type ship-related sources in Shanghai are shown in Table 3. The emissions of SO_2, NO_X, and $PM_{2.5}$, and VOC_s from inland-water ships and coastal ships accounted for the majority of primary emissions from all shipping related sources in Shanghai port, ranging from 72% for VOCs to about 99% for SO_2. They comprised about 17.4% of SO_2, 24.5% of NO_x, 5.2% of $PM_{2.5}$ and 0.6% of VOC_s emissions from all pollution sources in Shanghai. The shipping emissions in Shanghai port were estimated to account for 23% of SO_2, 26% of NO_x, and 23% of $PM_{2.5}$, and 28% of VOC_s from total shipping emissions in YRD.

Emissions estimates from this study fall within the range of estimates from Fu et al.'s study conducted in 2010 for the same region other studies (Fu et al., 2012; Fu et al., 2017). On the basis of shipping visa data, Fu et al. (2012) determined that the total amounts of SO_2, NO_X, and $PM_{2.5}$ in the vicinity of Shanghai port in 2010 were 3.5×10^4 ton/yr, 4.7×10^4 ton/yr, and 3.7×10^3 ton/yr, respectively, substantially lower than estimates in ~~our~~ this study. Using AIS data, Fu et al. (2017) reported 5 × 10^4 tons of SO_2 and 7× 10^4 tons of NOx from shipping in Shanghai port in 2013, close or a bit lower than the results in this study.

Within Shanghai, following the geographical division, inland-water ships were the most important ship-related source of emissions, accounting for 67% of SO_2, 66% of NO_x, 62% of $PM_{2.5}$ and 57% of VOC emissions from all ship-related sources in Shanghai (Table 2). ~~They comprised about 12% of SO_2, 19% of NO_x, 4% of $PM_{2.5}$ and 0.5% of VOC emissions from all pollution sources in Shanghai.~~ Emissions of SO_2, NO_X, $PM_{2.5}$ and VOCs from cargo trucks and port terminal equipment comprised a smaller percentage of emissions from all shipping related sources and particularly from all pollution sources so were therefore combined into one category in model simulation.

The spatial patterns of annual emissions from ship-related sources in Shanghai are shown using SO_2 as an example in Fig. ~~3c~~ 2c and Fig. ~~3d~~2d. SO_2 emissions from coastal ships were more prominent on the east-west shipping lanes and the vicinity of Yangshan port (Fig. ~~3c~~2c) while SO_2 emissions from inland water-going ships were significant concentrated along the Yangtze River and the Huangpu River, which run through the center of Shanghai.

3.2 The impact of shipping emissions on air quality

3.2.1 Contribution to ambient concentrations of SO_2 and $PM_{2.5}$ from all ships in YRD

On average, ships contributed 0.55 μg/m^3 in January (Fig. ~~4a~~3a) and 0.73 μg/m^3 in June (Fig. ~~4c~~3c) to the land ambient SO_2. The contribution of shipping emissions to the ambient monthly-average SO_2 concentration was higher in June 2015 than in January 2015 in the YRD region. The contribution from ships to land ambient SO_2 concentration peaked at 6.0 μg/m^3 (24.3% of ambient SO_2) in January and 8.84 μg/m^3 (69.7% of ambient SO_2 from all pollution sources) in June.

On average, ships contributed 0.36 μg/m^3 in January (Fig. ~~4b~~3b) and 0.75 μg/m^3 in June (Fig. ~~4d~~3d) to the ambient $PM_{2.5}$ concentrations across the YRD. Similarly, the contribution of shipping emissions to ambient monthly-average $PM_{2.5}$ concentrations was higher in June 2015 than in January 2015 in the YRD region. The contribution from ships to ambient $PM_{2.5}$ concentration peaked at 1.84 μg/m^3 (2.2% of the total ambient $PM_{2.5}$ concentration from all pollution sources) in January and 4.62 μg/m^3 (18.9 % of total ambient $PM_{2.5}$) in June. The highest shipping contributions to $PM_{2.5}$ were located near the Shanghai port.

The differences between January and June contributions of shipping to air quality mainly reflect differences in meteorology. The summer monsoon winds in June flow from the sea toward and, transporting shipping emissions inland in June whereas the winter monsoon winds in January transport shipping emissions out to sea. Differences in shipping emissions did not explain the different results for January and June. Monthly shipping emissions in YRD were 1.9×10^4 tons of SO_2 and 2.3×10^3 tons of $PM_{2.5}$ in January and 1.8×10^4 tons of SO_2 and 2.3×10^3 tons of $PM_{2.5}$ in June.

3.2.2 The influence of different offshore coastal areas in YRD on air quality

Shipping emissions on inland waters or within 12 NM of shore accounted for 30% to 85% of the total air quality impacts of ships within 200 NM of shore in January and June 2015, respectively (Fig. ~~5~~4). These results are similar to those of Lv et al. (2018) who reported that shipping emissions within 12 NM of shore contributed 30% to 90% of the $PM_{2.5}$ induced by emissions within 200 NM. On average, ships contributed 0.24 μg/m^3 to the ambient $PM_{2.5}$ in January (Fig.~~5a~~4a) and 0.56 μg/m^3 to ambient $PM_{2.5}$ concentrations in June (Fig.~~5f~~4f). Peak contributions were 1.62 μg/m^3 $PM_{2.5}$ in January and 4.02 μg/m^3 $PM_{2.5}$ in June, respectively.

The average and peak contributions from the shipping emissions in specific offshore coastal areas to the ambient SO_2 and $PM_{2.5}$ concentrations on shore for the two months are listed in Table S4. Shipping emissions beyond 12 NM had a much smaller impact on ambient SO_2, which average contributions were below 0.01 μg/m^3 and peak contributions were below 0.06 μg/m^3 (Table S4).

Shipping emissions at distances of 12-24 NM, 24-48 NM and 48-96 NM from shore contributed on average 0.01-0.07 μg/m^3 to the ambient $PM_{2.5}$ concentrations. Peak contributions of shipping emissions from areas beyond 12 NM ranged from 0.05 μg/m^3 (12-24 NM) to 0.14 μg/m^3 (48-96 NM) in January (Fig. ~~5b~~4b-d); the peak influence was higher in June and ranged from 0.2 μg/m^3 (12-24 NM) to 0.34 μg/m^3 (24-48 NM) (Fig. ~~5g~~4g-i). In the YRD region, shipping emissions on inland waters or within 12 NM of shore had larger contributions to ambient $PM_{2.5}$ than did more distant ships~~ (Fig. 5). However~~, but the busy north-south shipping lanes in the distant region from shore also impacted ambient $PM_{2.5}$ concentrations. Shipping emissions from 96 to 200 NM from shore had little impact on air quality over land and contributed less than 0.05 μg/m^3 (or 3% of the ship-related contribution) to the ambient land $PM_{2.5}$ (Fig. 5e 4e and Fig. 5i4i).

The cumulative contributions to ambient SO_2 concentrations in the 16 core YRD cities from ships at different distances from shore in January and June 2015 differed from $PM_{2.5}$ results (Fig. 65). In both January (Fig. 6a5a) and June (Fig. 6c5c), shipping emissions within 12 NM accounted for at least 78% of the ship-related contribution to ambient SO_2 concentrations in these cities. Shipping emissions beyond 12 NM had limited contribution to SO_2 concentrations in 16 core YRD cities, implying that the boundary of 12 NM might be suitable for regulating SO_2 emissions. This could also be proved by Schembari et al., (2012), who reported that statistically significant reductions of SO_2 levels (66% to 75%) were found in 3 out of the 4 European harbours, 5 months after the implementation of the EU directive 2005/33/EC that requires all ships at berth or anchorage in European harbours use fuels with a sulfur content of less than 0.1% from January 2010. The quicker chemical reaction and shorter lifetime of SO_2 may explain why ships further out than 12 NM had much smaller impact on land ambient SO_2 concentrations (Collins et al., 2009; Krotkov et al., 2016). SO_2 reacts under tropospheric conditions via both gas-phase processes (with OH) and aqueous-phase processes (with O_3 or H_2O_2) to form sulfate aerosols, and is also removed physically via dry and wet deposition (Seinfeld and Pandis, 2006). The sulfur deposition due to shipping emissions is mainly contributed by the dry depositions (Chen et al.,2019). In the Planet boundary layer (PBL), SO_2 has short lifetimes (less than 1 day during the warm season) and are concentrated near their emission sources (Krotkov et al., 2016).which may be attributed to the quicker chemical reaction and shorter lifetime of SO_2 (Junkermann and Roedel, 1983).

In contrast to SO_2, the cumulative contributions to $PM_{2.5}$ in the 16 core YRD cities from ships at different distances from shore showed greater differences in January and June 2015. In January, the relative contributions of ships inland or within 12 NM of shore to ship-related $PM_{2.5}$ concentrations ranged from 78.7% in Zhoushan, which were mostly influenced by the closest shipping emissions, to 26.3% in Yangzhou (Fig. 6b5b). In June, the relative contributions of ships inland or within 12 NM of shore to all $PM_{2.5}$ emissions from ships ranged from 85.2% in Nanjing to 54.6% in Taizhou (Fig. 6d5d). Therefore, in both months, shipping emissions within 12 NM were a major contributor to ship-related $PM_{2.5}$ concentrations in most of core YRD cities.

Although busy north-south shipping lanes 24-96 NM from shore contributed little SO_2 concentrations to YRD cities, shipping emissions from this area contributed 12% to 39% of ship-related $PM_{2.5}$ concentrations in YRD cities. Of $PM_{2.5}$ in YRD cities contributed by ships within 200 NM of shore, 97% is accounted for by shipping emissions within 96 NM of shore. The results of these YRD analyses suggest that although ambient ship-related SO_2 concentrations were mainly affected by shipping inland or within 12 NM, expanding China's current DECA to around 100 NM or more would reduce the majority of the impacts of shipping on regional $PM_{2.5}$ pollution. It also implies that the future ECA policy should consider multiple air pollutants including the primary and secondary pollutants synchronically.

3.2.3 The influence of different ship-related sources in Shanghai port on air quality

The impact of port-scale shipping-related sources on ~~city-scale~~the air quality in Shanghai was significant, and the dominant sources of shipping-related emissions (i.e., coastal ships, inland-water ships, and other shipping-related sources) varied depending on the season and their locations relative to cities (Figure ~~7~~6). Inland-water ships had a larger influence on areas within Shanghai near the Yangtze River and Huangpu River. Inland-water ships contributed on average 0.24 μg/m^3 in January (Fig.~~7a~~6a) and 0.37 μg/m^3 in June (Fig.~~7d~~6d) to ambient $PM_{2.5}$, and accounted for 40% to 80% of all $PM_{2.5}$ from ship-related sources. The inland-water ships had their large influence in areas near the cross section of Yangtze River and Huangpu River, where their contributions to ambient $PM_{2.5}$ peaked at 1.87 μg/m^3 in January and 2.67 μg/m^3 in June (Fig.~~7a~~ 6a and Fig. ~~7d~~6d). Coastal ships contributed on average 0.02 μg/m^3 in January and 0.30 μg/m^3 in June to ambient land $PM_{2.5}$ concentrations. Peak contributions of coastal ships to ambient $PM_{2.5}$ were 0.1 μg/m^3 in January (Fig.~~7b~~6b) and 0.71 μg/m^3 in June (Fig.~~7e~~6e). The impact of coastal ships was much smaller in January than in June due to meteorological reasons described earlier. Container-cargo trucks and port terminal equipment contributed on average 0.15 μg/m^3 in January (Fig.~~7c~~6c) and 0.12 μg/m^3 in June (Fig.~~7f~~6f) to ambient $PM_{2.5}$ concentrations, and accounted for 10 to 45% of $PM_{2.5}$ from shipping-related sources. Peak contributions of container-cargo trucks and port terminal equipment were 2.14 μg/m^3 in January and 1.40 μg/m^3 in June. The slightly larger contribution of container-cargo trucks and terminal equipment to $PM_{2.5}$ concentrations was mainly because the lower wind speed in winter hindered the dispersion of pollutants. Although the contributions of container-cargo trucks and port terminal equipment to ambient $PM_{2.5}$ were generally lower than the contributions of ships, these other shipping-related sources were still important in both winter and summer due to their impact on air quality near the Shanghai city center.

3.3 Population-weighted $PM_{2.5}$ concentrations

3.3.1 Influence of different offshore coastal areas in YRD on population-weighted $PM_{2.5}$

Population-weighted $PM_{2.5}$ concentrations in the YRD from shipping-related sources were larger in June (0.4 μg/m^3 to 2.6 μg/m^3 in June; Fig. 8d7d) than in January (0.1 μg/m^3 to 1.2 μg/m^3; Fig. 8b7b). This is in contrast to population-weighted $PM_{2.5}$ concentrations from all pollution sources, which were higher in January (33.1 μg/m^3 to 80.2 μg/m^3; Fig. 8a7a) than in June (9.5 μg/m^3 to 48.4 μg/m^3; Fig. 8c7c). Thus, population-weighted $PM_{2.5}$ concentrations from shipping sources accounted for 0.9% to 15.5% of the population-weighted $PM_{2.5}$ concentrations from all pollution sources in June, larger than the contributions of 0.2% to 1.6% in January, which was attribute to higher shipping-related population-weighted $PM_{2.5}$ concentrations in June and higher population-weighted $PM_{2.5}$ concentrations from all pollution sources in January. Of the 16 core YRD cities, the highest ship-related population-weighted $PM_{2.5}$ concentrations were found for Shanghai in June (2.6 μg/m^3), 1.5 times higher than the second-highest city Nantong (1.7 μg/m^3). The six cities in the YRD with the largest contributions of $PM_{2.5}$ from shipping sources were all coastal cities, which suggests as expected that people living in coastal regions would have higher exposures to air pollution from shipping-related sources than people living in farther inland, especially during the summer monsoon.

Taking the population-weighted $PM_{2.5}$ concentrations from all shipping sources within 200NM as the base, the shipping, both in inland waters and within 12NM of shore, was a major contributor to population-weighted $PM_{2.5}$ concentrations in 16 YRD cities; they accounted for 52.9% to 82.7% (Fig. 7e). The Population-weighted $PM_{2.5}$ concentrations from shipping within 12-24NM from shore were much smaller, accounting for 2.5% to 6.6%. But shipping emissions in the area24-48 NM accounted for 6.8% to 11.5% and ships 48-96 NM from shore accounted for 6.3%

to 31.6%. These contributions in greater distance were larger than the contribution from ships in 12-24 NM from shore, probably because the busier shipping lanes fall within the more remote areas like 24-48 NM from shore. Therefore, although shipping inland and within 12 NM of shore was the dominant contributor to potential population exposure to $PM_{2.5}$, ships as far as 24-96 NM could also be important.

3.2.3 3.3.2 The influence of different ship-related sources in Shanghai port on potential exposure

Of the shipping-related sources in Shanghai, inland-water ships were the largest contributors to both $PM_{2.5}$ and population-weighted $PM_{2.5}$ (Fig. 9b8b). The population-weighted $PM_{2.5}$ in January was 0.38 μg/m^3 from inland-water ships (Fig. 9a8a). In June, the population-weighted $PM_{2.5}$ from inland-water ships contributed reached 0.57 μg/m^3 because the region near the Huangpu River and Yangtze River had a high population where inland-water ships contributed high levels of $PM_{2.5}$ (Fig. 9b8b). In contrast, coastal ships contributed 0.27 μg/m^3 and container-cargo trucks and port terminal equipment contributed only 0.14 μg/m^3 to population-weighted $PM_{2.5}$ in June. Population-weighted $PM_{2.5}$ from shipping sectors in January were lower than those in June, while population-weighted $PM_{2.5}$ from container-cargo trucks and port terminal equipment was slightly higher. In both June and January, population-weighted $PM_{2.5}$ concentrations from ship-related sources were larger than the average $PM_{2.5}$ concentrations from ship-related sources because the population was denser in the areas most highly influenced by shipping-related sources (Fig. 9a 8a and 9b8b). The difference between average $PM_{2.5}$ concentration and population-weighted $PM_{2.5}$ concentration was largest for inland-water ships, which contributed two times more population-weighted $PM_{2.5}$ concentration than the average $PM_{2.5}$ concentration.

Population-weighted $PM_{2.5}$ concentrations were not evenly distributed among the 16 administrative districts in Shanghai. The population-weighted $PM_{2.5}$ from all pollution sources ranged from 44.8 μg/m^3 to 124.5 μg/m^3 in January (Fig. 9c8c) and 23.4 μg/m^3 to 67.2 μg/m^3 in June (and Fig. 9g8g). Heavy motor vehicle traffic probably contributed to higher population-weighted $PM_{2.5}$ in the city center (Huangpu, Jingan and Hongkou).

Areas in the city center had high population-weighted $PM_{2.5}$ from inland-water ships because of the combination of dense population and location close to Huangpu River (Fig. ~~9d~~ 8d and Fig. ~~9h~~8h). Among them, Baoshan and Yangpu had the highest population-weighted $PM_{2.5}$ concentrations from inland-water ships (both around 1.31 μg/m^3) in June. Besides, in June, population-weighted $PM_{2.5}$ from coastal ships ranged from 0.17 μg/m^3 to 0.40 μg/m^3, and the coastal district (Fengxian) suffered the largest impacts. Transport of emissions by the summer monsoon caused impacts on population-weighted $PM_{2.5}$ not only in coastal districts but also in the highly populated city center. As for population-weighted $PM_{2.5}$ caused by container-cargo trucks and port terminal equipment, Baoshan had the highest population-weighted $PM_{2.5}$ in both January (0.4 μg/m^3) and June (0.45 μg/m^3) due to its high population and location close to the source (Fig. ~~9f~~ 8f and ~~9j~~8j). The results of the analyses of different-type shipping-related sources indicated that ship-related sources close to densely-populated areas contribute substantially to population exposures to air pollution

3.4 Limitations and uncertainties

Limitations in the study were mainly related to some missing information in database and assumptions during estimation of shipping emissions. When estimating shipping emission inventory, underestimations of actual emissions may be introduced by missing information. For example, AIS data has a high coverage of coastal vessels, but many inland vessels are not equipped with AIS. Emissions from those inland vessels without AIS devices were supplemented by using 2015 vessel call data provided by Shanghai MSA and Shanghai Municipal MSA, and that could introduce some uncertainties for inland river vessels. Also, emissions from fishing boats were probably underestimated because AIS devices on some fishing boats may not be in use. Similarly, limited information exists on auxiliary boilers in the Lloyd's register and CCS databases so we calculated the main engine and auxiliary engine emissions but did not consider auxiliary boiler emissions in this study, which may cause underestimation of shipping emissions.

In addition, we did not consider the external effects of water flow, wind, and waves when calculating engine power for ships going over the region. This would introduce some uncertainties (Aulinger et al., 2016). According to the previous studies in other areas, these factors may increase fuel consumption of individual vessels by as much as 10% to 20%, while the effects of waves on emissions estimations over extensive geographical regions are negligible (Jalkanen et al., 2009; Jalkanen et al., 2012). The downstream of the Yangtze River is located in the geographically plateau region, and the river flow is below 0.5 m/s (Song and Tian, 1997; Xue et al., 2004). For Shanghai, located at the end of mouth of the Yangtze River to the East China Sea with a flat terrain, the river flow is very slow. Given that ships traveling the Yangtze River near Shanghai have speeds over ground (SOG) of about 5-10 knots (3-5 m/s), the relative ratios of water flow to the SOG is within 20%. In our future work, we will fill the gap in the basic ship data and consider the external effects when building the shipping emission inventory.

Finally, this work only extends from emissions to air quality and population exposures. The health impacts of shipping-related air pollution in Shanghai and the YRD region will be explored in future work.

4 Conclusions

As the major economic and shipping center in China, the YRD, and in particular Shanghai, experiences high emissions of shipping-related pollutants that result in significant contributions to ambient and population-weighted air pollutant concentrations. Our results showed that on average in 2015 ships contributed 0.75 μg/m^3 to the ambient land $PM_{2.5}$ in YRD, with a peak of 4.62 μg/m^3 (18.9% of the total ambient $PM_{2.5}$ concentration from all pollution sources) near Shanghai Port. The shipping emissions affecting air quality in the YRD were mainly within 12 NM of shore (over 75% for ship-related SO_2 and 50% for ship-related $PM_{2.5}$ concentrations) but emissions coming from 24 to 96 nm offshore also contributed substantially to $PM_{2.5}$ concentrations in the YRD under the transport of summer monsoon. The megacities of Shanghai and Nantong had the highest ship-related population-weighted $PM_{2.5}$ concentrations from the combination of high population density and high shipping emissions. In Shanghai, the inland-water ships contributed a majority (40-80%) of the $PM_{2.5}$ from shipping-related sources; inland-water ships also contributed prominently to population-weighted $PM_{2.5}$ in several districts in Shanghai. These study results on contributions of ships at different distances from shore in the YRD and shipping-related sources in and near Shanghai to ambient air quality and population-weighted $PM_{2.5}$ could inform future ECA policies. For example, policymakers could consider whether to expand China's current DECA boundary of 12 NM to around 100 NM or more to reduce the majority of shipping impacts on air pollution concentrations and exposure. It will be helpful to improve the local air quality and reduce human exposures in densely populated areas by developing more stringent regulations on the fuel quality for ~~both coastal and inland water~~ ships entering inland rivers or other waterways close to residential regions.

Author contribution

YZ and KW conceived the study and made a roadmap for organizing this paper. JF did the air quality simulation and wrote the manuscript. SL ran the shipping emission inventory model. JM ran the WRF model. YZ and CH provided port-related emission inventory. CL and HK provided roadmap for human exposure analysis. AP and WM provided constructive comments in analyzing data. JA and LL provided local-scale land-based emission inventory. YS and JL provided river shipping emission data. XW and QF provided monitoring data. SW and DD provided national land-based emission inventory. JC, WG, and HZ provided container cargo-car traffic emission inventory.

Acknowledgement

This research work was partly supported by the National Key Research and Development Program of China (grant no. 2016YFA060130X) and the National Natural Science Foundation of China (21677038). Katherine Walker and Allison Patton give their thanks to the Bloomberg Foundation. The authors wish to thank Noelle Selin and Freda Fung for providing valuable advice. The opinions expressed in this article are the author's own and do not reflect the view of Fudan University, Shanghai Academy of Environmental Science, Shanghai Environmental Monitoring Center, Tsinghua University, Shanghai Urban-rural Construction and Transportation Development Research Institute, or the Health Effects Institute or its sponsors, nor do they necessarily reflect the views and policies of the EPA or motor vehicle and engine manufacturers.

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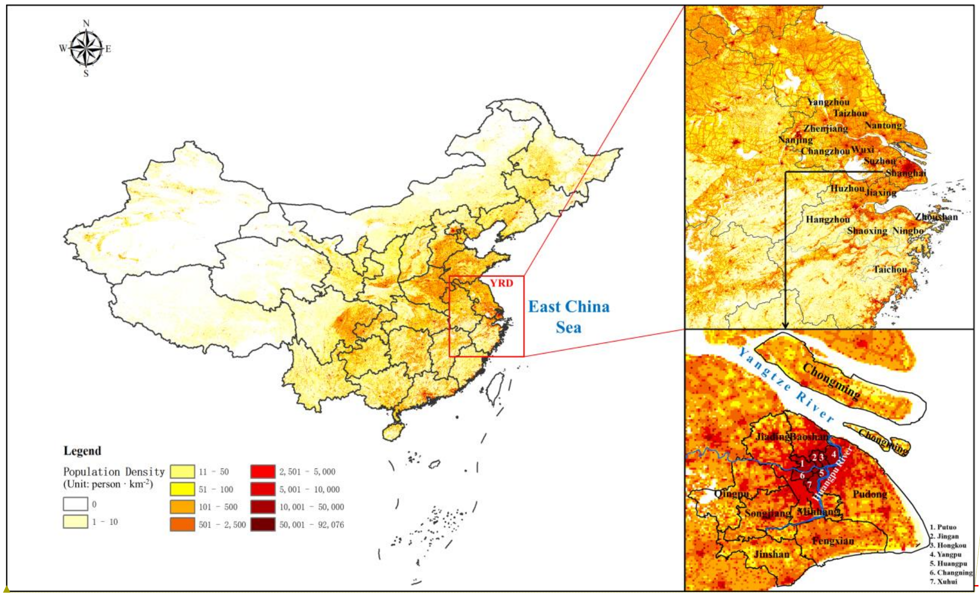

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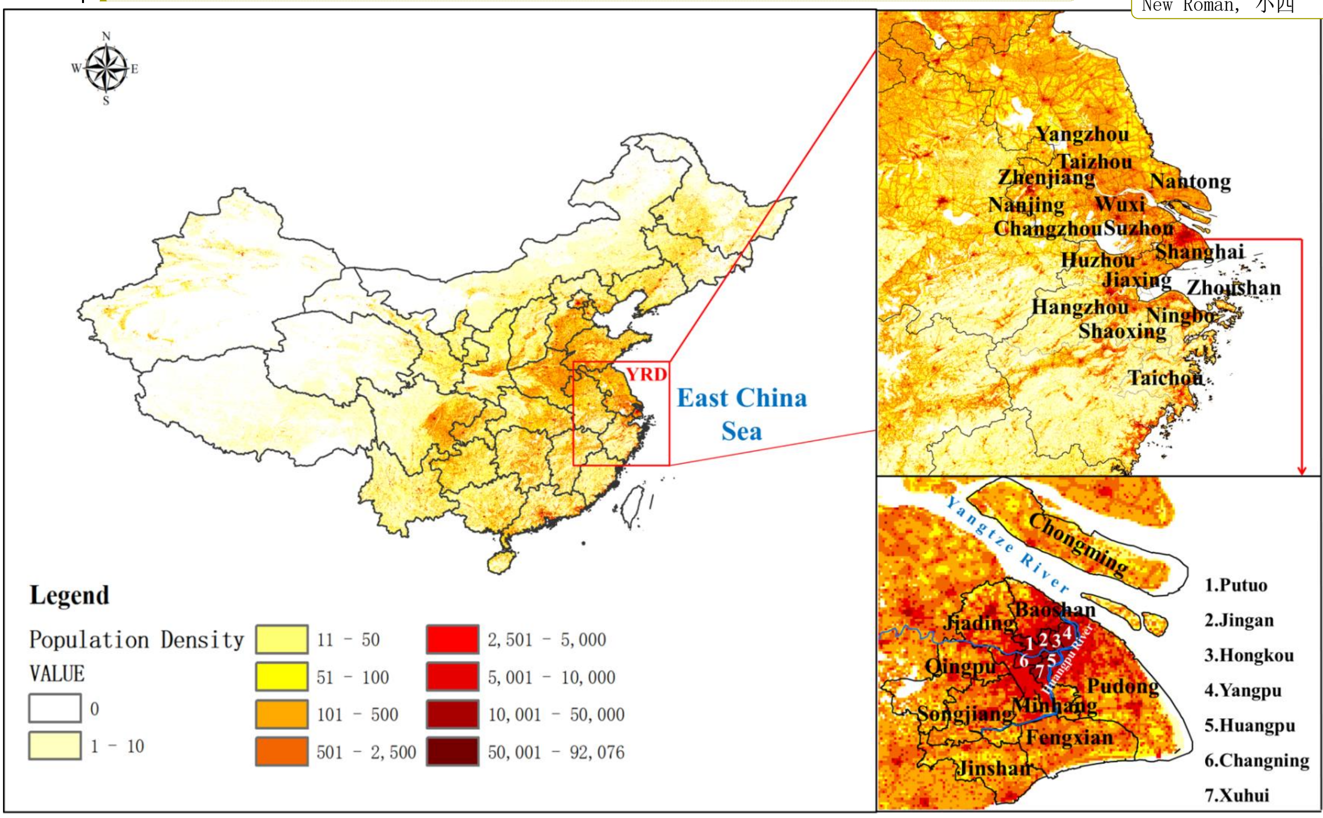


Figure 1. Geographic location of the study area YRD/Shanghai with population density in 2015. 16 core cities in YRD and 16 administrative districts in Shanghai are

noted on the map. The smaller administrative districts are labeled with numbers: Putuo (1), Jingan (2), Hongkou (3), Yangpu (4), Huangpu (5), Changning (6), Xuhui (7).

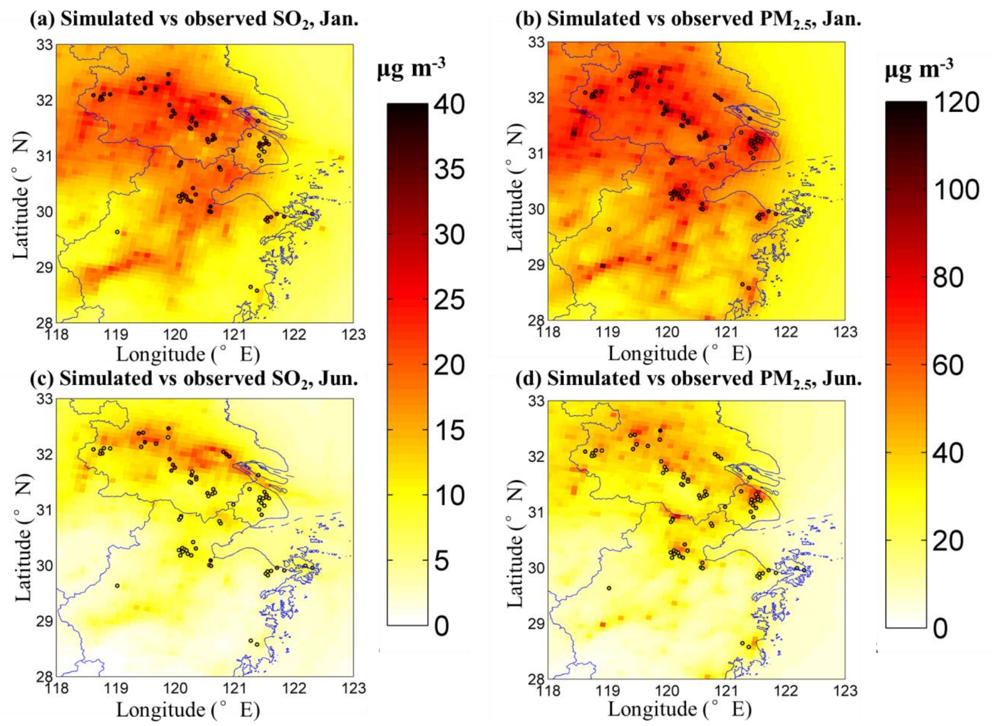
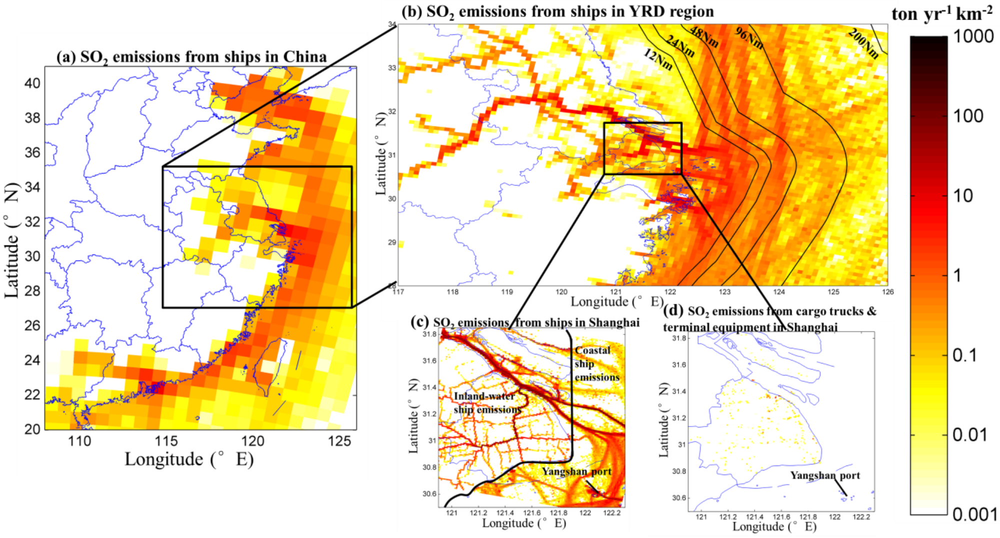


Figure 2. The simulated (grid) and observed (circles) SO_2 concentration distribution in YRD region, in January 2015 (a) and June 2015 (c); the simulated (grid) and observed (circles) $PM_{2.5}$ concentration distribution in YRD region, in January 2015 (b) and June 2015 (d)



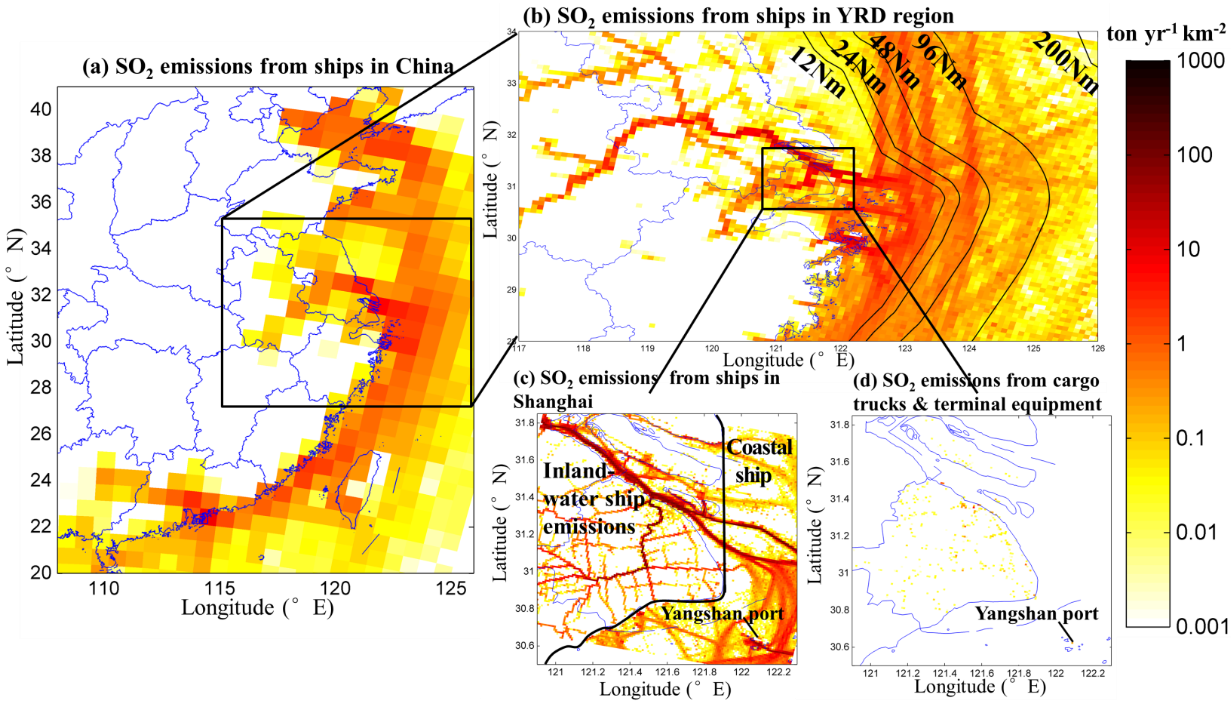


Figure ~~3~~2. SO_2 emissions in 2015 from (a) shipping traffic in China (the average value of January and June) at resolution of 81km × 81km; (b) ships in different offshore coastal areas (inland-water and within 12 NM, 12-24 NM, 24-48 NM, 48-96 NM and 96-200 NM) in the YRD region, at resolution of 9km × 9km; (c) inland-water ships and coastal ships in Shanghai, at resolution of 1km × 1km; and (d) container-cargo trucks and port terminal equipment in Shanghai, at resolution of 1km × 1km. The black line in (c) refers to the division line between the inland water and coastal water for Megacity Shanghai defined in this study.

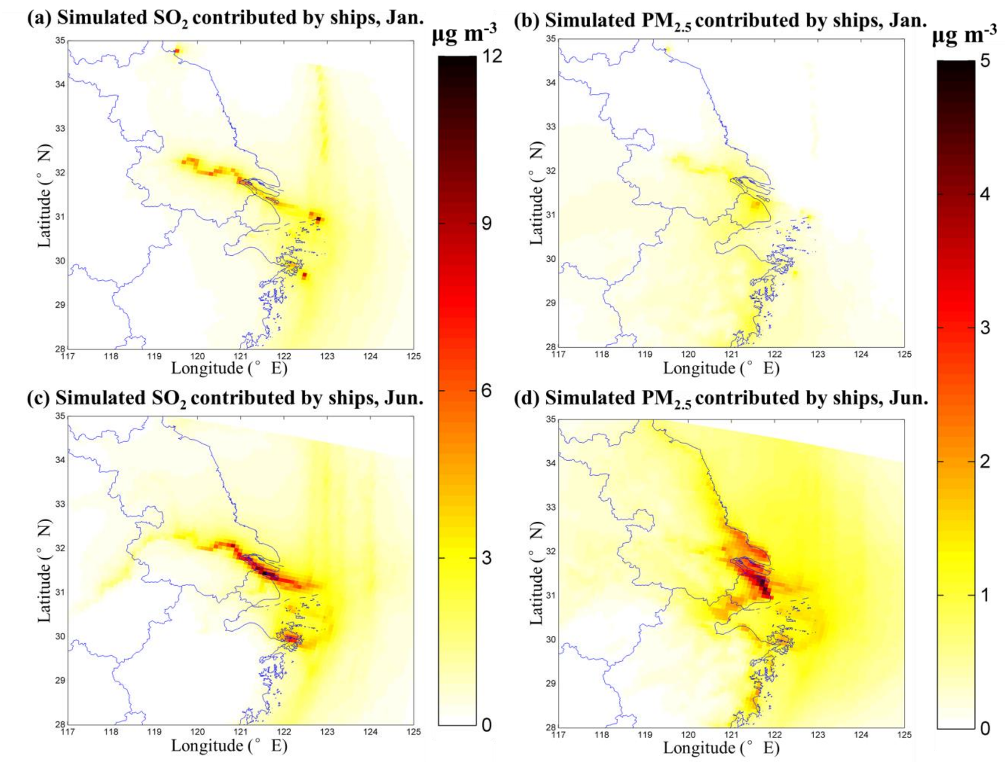


Figure 43. Simulated SO_2 (a, c) and $PM_{2.5}$ (b, d) concentrations contributed by shipping traffic sources in YRD region, in January 2015 (a, b) and June 2015 (c, d)

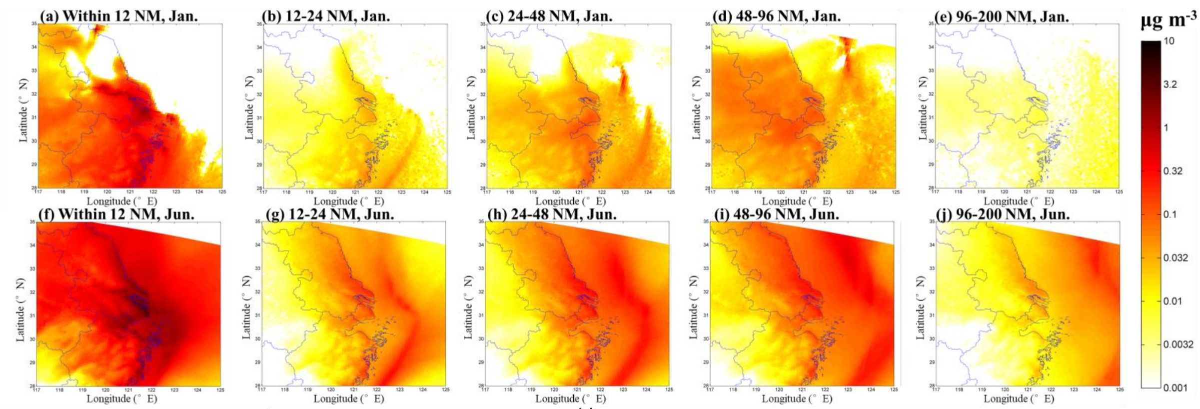


Figure ~~5~~4. Contributions to $PM_{2.5}$ concentrations from shipping emissions at distances within 12 NM of shore (including inland-waters) (a, f), 12 to 24 NM from shore (b, g), 24 to 48 NM from shore (c, h), 48 to 96 NM from shore (d, i) and 96 to 200 NM from shore in January 2015 (a-e) and in June 2015 (f-j).

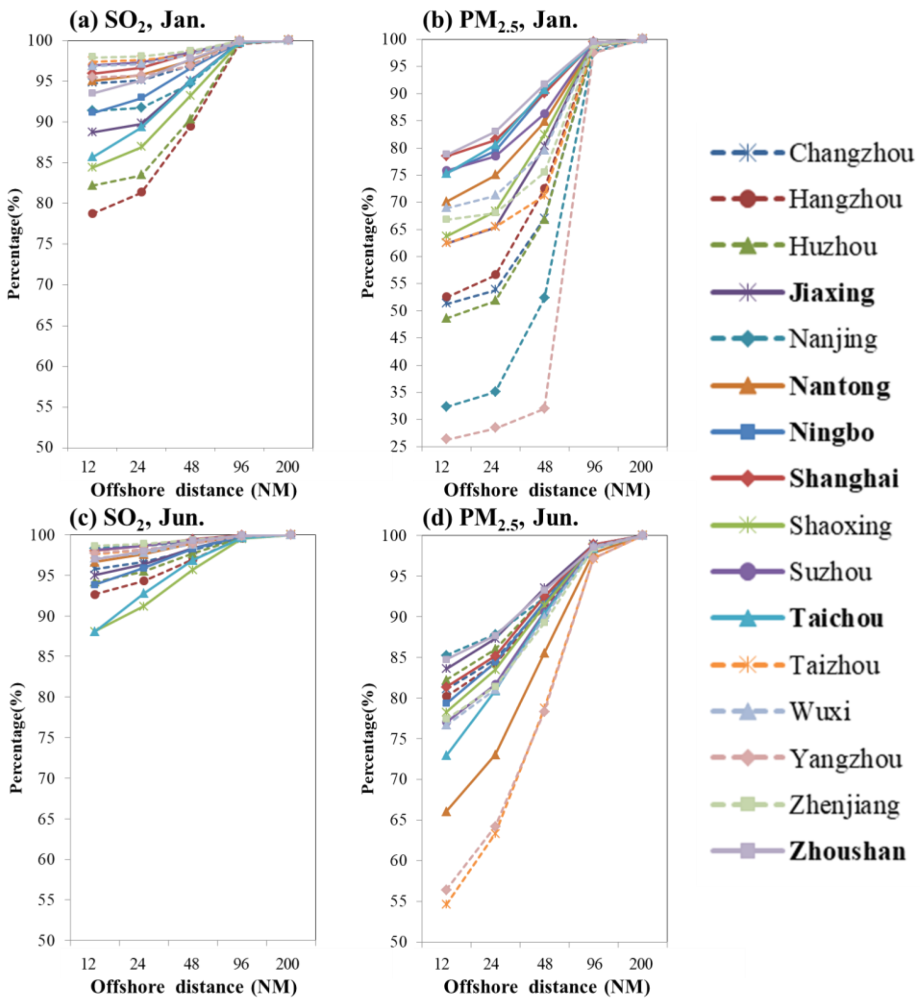


Figure ~~6~~5. Cumulative contributions of shipping emissions in the YRD at distances within 12 NM of shore (including inland-waters), 24 NM from shore, 48 NM from shore, 96 NM from shore, and 200 NM from shore to $PM_{2.5}$ concentrations (a, c) and SO_2 concentrations (b, d) in January 2015 (a, b) and in June 2015 (c, d). Names of Coastal cities are bold in the legend.

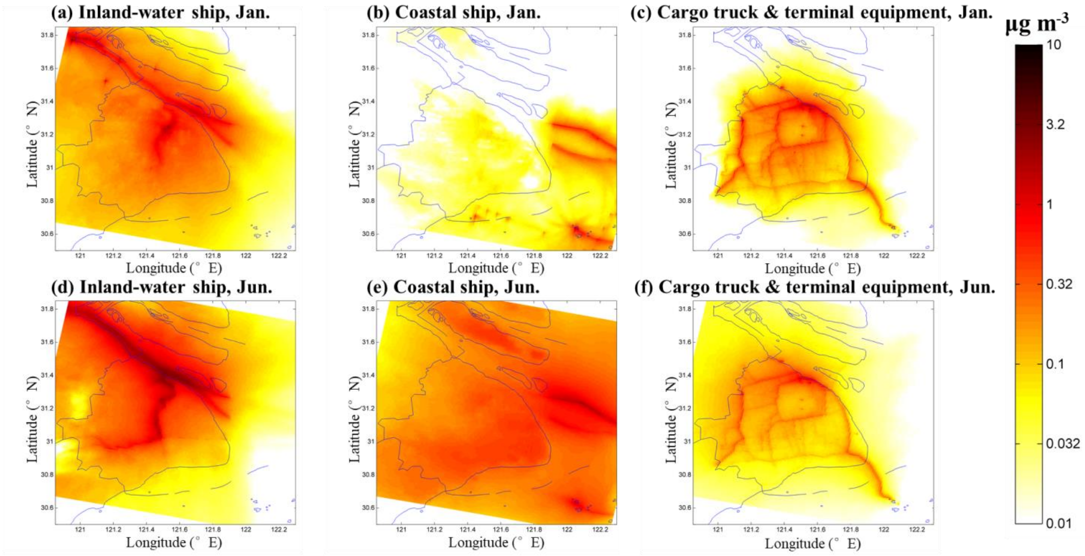


Figure ~~7~~6. Contributions to $PM_{2.5}$ concentrations from inland-water ships (a, d), coastal ships (b, e) and container-cargo trucks and port terminal equipment (c, f) in January 2015 (a-c) and June 2015 (d-f).

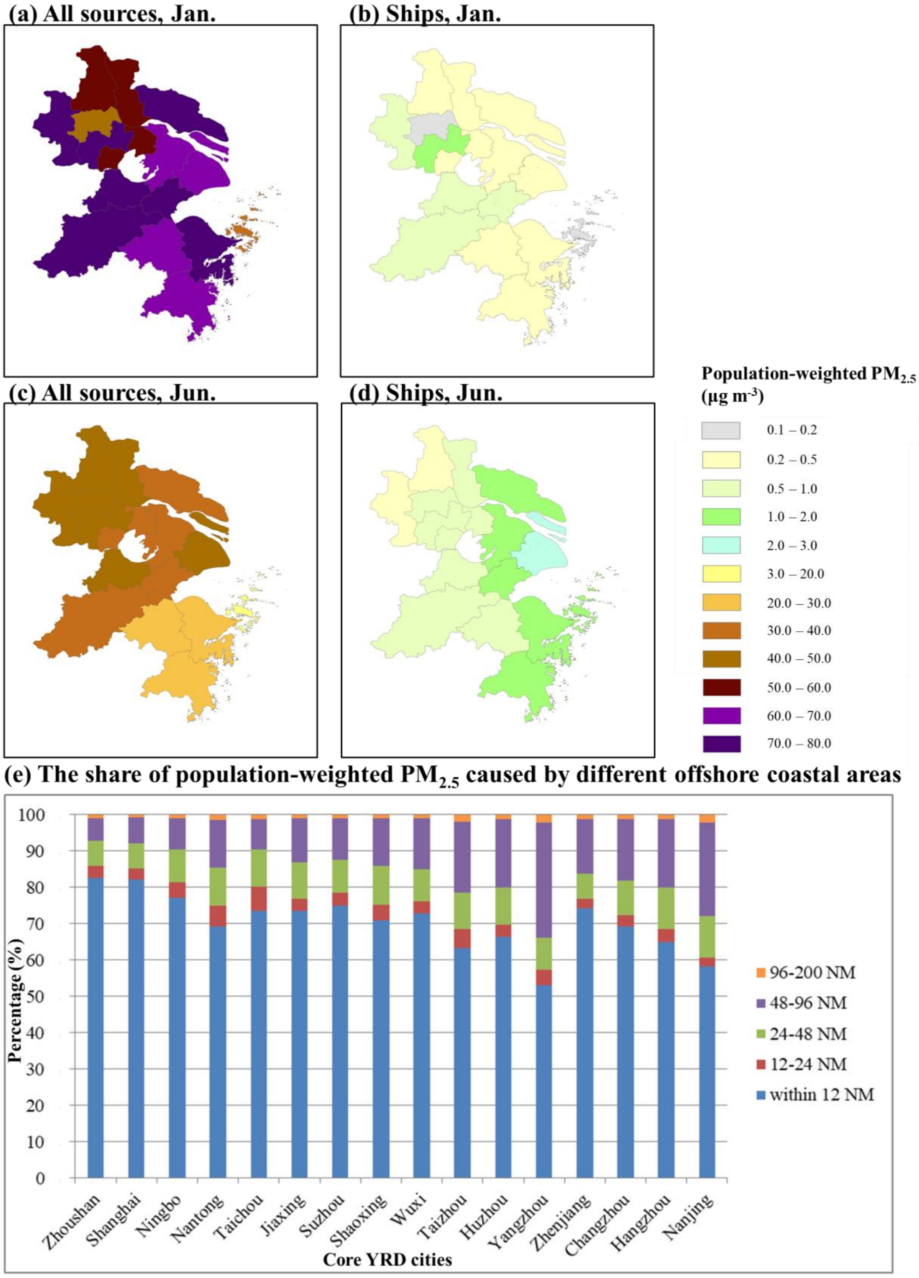
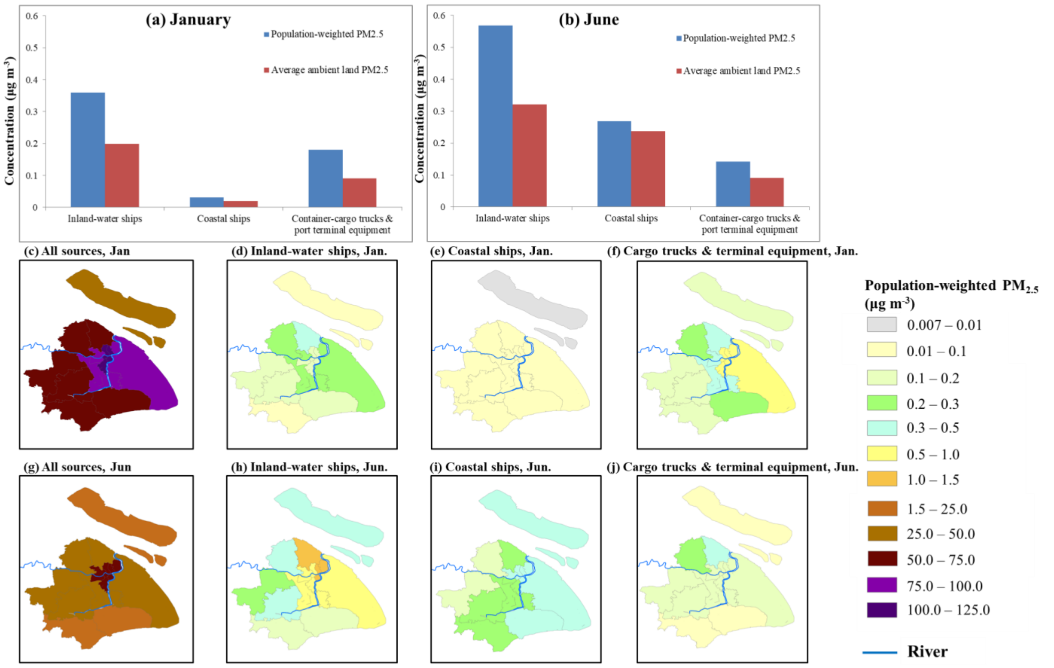


Figure 87. The spatial distribution of population-weighted $PM_{2.5}$ in 16 YRD cities caused by all pollution sources (a, c) and by all ships (b, d) in January 2015 (a, b) and June 2015 (c, d); the average share of population-weighted $PM_{2.5}$ in 16 YRD cities caused by different offshore coastal areas in all ships (e). The cities' names are ordered by their distance to the coast.



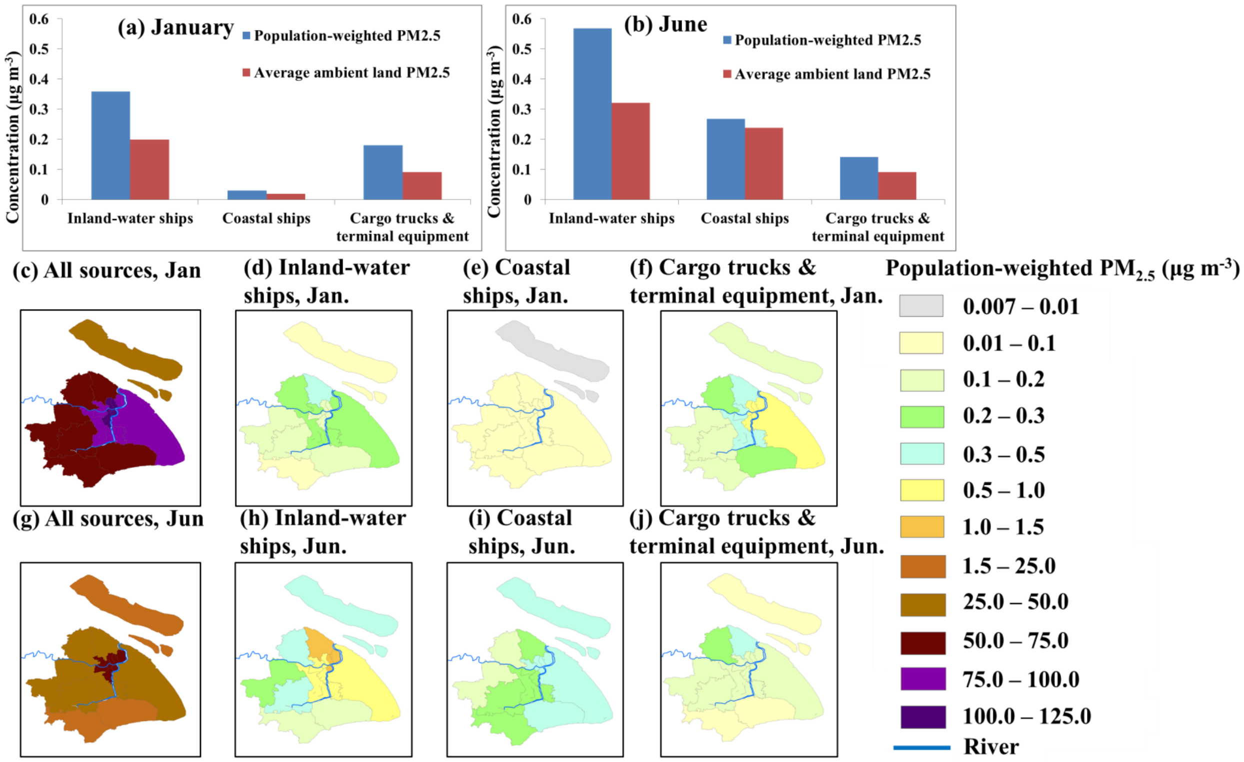


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Figure ~~9~~8. Population-weighted $PM_{2.5}$ and average $PM_{2.5}$ caused by different ship-related sources in Shanghai, in January(a) and in June (b); population-weighted $PM_{2.5}$ caused by all pollution sources (c, g), inland-water ships (d, h), coastal ships (e, i) and container-cargo trucks and port terminal equipment (f, j) in 16 districts in Shanghai, in January 2015(c-f) and June 2015 (g-j).

Table 1 Statistical metrics of the model evaluation. Observed data (Obs.) and simulated data (Sim.) for each city are the average of monthly values of January and June case. NMB, NME, RMSE and *r* were calculated based on the daily-average observed and simulated data.

City	SO_2						$PM_{2.5}$					
	Obs.	Sim.	NMB (%)	NME (%)	RMSE (μg m^{-3})	*r*	Obs.	Sim.	NMB (%)	NME (%)	RMSE (μg m^{-3})	*r*
Changzhou	31.24	20.14	-35.55	40.85	15.79	0.80	74.21	68.27	-8.01	32.51	31.99	0.76
Hangzhou	16.84	13.75	-18.35	28.74	6.77	0.83	59.35	56.96	-4.03	28.21	22.05	0.75
Huzhou	19.25	14.73	-23.52	38.81	11.45	0.80	65.13	70.50	8.25	45.60	39.17	0.47
Jiaxing	25.37	16.84	-33.67	50.58	17.31	0.75	61.31	57.01	-7.02	33.98	29.96	0.65
Nanjing	22.39	16.38	-20.60	26.50	10.13	0.76	68.20	55.71	-14.06	27.80	32.30	0.60
Nantong	32.73	22.05	-32.66	49.69	23.21	0.70	68.69	51.15	-25.54	39.27	37.23	0.69
Ningbo	16.20	10.47	-35.42	42.01	7.64	0.83	55.47	48.06	-13.37	34.51	28.49	0.75
Shanghai	19.16	12.32	-35.72	40.23	10.72	0.83	63.64	67.77	6.50	36.18	28.71	0.75
Shaoxing	22.47	14.63	-34.91	40.03	10.36	0.80	61.90	56.86	-8.15	34.06	27.21	0.70
Suzhou	21.37	15.16	-29.09	37.26	10.39	0.85	67.11	56.45	-15.89	33.41	28.76	0.76
Taichou	10.72	7.55	-29.64	34.07	5.25	0.80	47.55	43.69	-8.11	35.35	24.09	0.52
Taizhou	29.64	20.84	-29.70	61.53	22.63	0.67	74.56	62.82	-15.75	31.75	33.49	0.63
Wuxi	24.64	18.89	-23.35	30.85	10.58	0.87	73.45	59.36	-19.20	31.80	30.92	0.77
Yangzhou	25.78	18.75	-27.31	44.22	15.17	0.62	62.30	60.12	-3.50	46.10	37.08	0.57
Zhenjiang	29.65	21.50	-27.51	39.49	16.23	0.61	67.78	62.61	-7.63	33.88	30.31	0.59
Zhoushan	9.99	8.04	-19.60	40.42	6.73	0.64	30.13	19.81	-34.28	49.15	16.82	0.78

Table 2. Primary emissions (ton/yr), emission share in all shipping emissions (%) and emissions density (ton/yr/km^2) from shipping at different boundaries in YRD region[a] in 2015

	Pollutants	Within 12 NM	12-24 NM	24-48 NM	48-96 NM	96-200 NM
Shipping emission inventory (ton/yr)	SO_2	1.3×10^5	1.4×10^4	2.5×10^4	3.2×10^4	1.3×10^4
	NO_x	3.6×10^5	2.0×10^4	3.5×10^4	4.5×10^4	1.8×10^4
	$PM_{2.5}$	1.3×10^4	2.4×10^3	4.5×10^3	5.4×10^3	1.5×10^3
	VOC_s	7.9×10^3	8.3×10^2	1.3×10^3	1.5×10^3	3.0×10^2
Emission share in all shipping emission (%)	SO_2	61.4	6.4	11.4	14.9	5.8
	NO_x	75.0	4.1	7.4	9.6	3.9
	$PM_{2.5}$	48.4	9.0	16.9	20.2	5.5
	VOC_s	66.6	7.0	11.2	12.6	2.6
Emission density (ton/yr/km^2)	SO_2	0.66	0.54	0.49	0.33	0.06
	NO_x	1.74	0.86	0.77	0.51	0.08
	$PM_{2.5}$	0.08	0.06	0.06	0.04	0.01
	VOC	0.05	0.02	0.01	0.01	0.001

a. domain 3

Table 3. Primary emissions (ton/yr) and emission share in all pollution sources (%) from different-type ship-related sources in Shanghai[a] in 2015

	Ship-related source	SO_2	NO_x	$PM_{2.5}$	VOC
Emission inventory (ton/yr)	Inland-water ships[b]	3.3×10^4	9.2×10^4	0.40×10^4	0.27×10^4
	Coastal ships[c]	1.6×10^4	2.9×10^4	0.18×10^4	0.067×10^4
	Container-cargo trucks	0.0	1.8×10^4	0.064×10^4	0.11×10^4
	Port terminal equipment[d]	0.0021×10^4	0.18×10^4	0.0057×10^4	0.022×10^4
Emission share in all pollution sources in Shanghai (%)	Inland-water ships	11.8	18.7	3.6	0.5
	Coastal ships	5.6	5.8	1.6	0.1
	Container-cargo trucks	0.0	3.7	0.6	0.2
	Port terminal equipment	0.01	0.36	0.05	0.04

a. domain 4

b. defined as ships operate in both the outer port and in the inner river region of Shanghai Port, which include Yangtze River, Huangpu River and other river ways in Shanghai

c. includes China coastal and international ships

d. includes cranes and forklifts used for internal transport