

Response to Reviewer #1's Comments

General comment

The paper treats of ozone formation trend (2016-2020) due to shipping emission in China by using modelling simulations suggesting the relevance of this source on this pollutant. The topic is interesting and suitable for the Journal. However, some aspects related to the choice done in modelling and to the interpretation of results are not completely clear or well described, see my specific comments. For this reason, I suggest considering the paper for publication after a revision step.

Response:

Thank you for your overall assessment and constructive suggestions. We appreciate your recognition of the relevance and timeliness of our study. In response to your comments, we have carefully revised the manuscript to clarify the modeling choices, refine the interpretation of the results, and address the specific concerns you raised. We hope the updated version more clearly conveys the scientific rationale, methodological robustness, and policy relevance of our work.

Specific comments

Comment 1

Anthropogenic emissions from other countries within the modeling domain (Table S2) was taken at 2010. It is possible to have a relevant uncertainty from this considering the period span of the study (2016-2020)?

Response:

Thank you for pointing out this important issue. We acknowledge that the use of anthropogenic emissions from other countries for the year 2010 could indeed introduce some uncertainty, particularly in boundary areas or regions with strong cross-border transport.

However, our primary focus is on the impacts of domestic shipping emissions within China, and most of the key regions of interest, such as the Yangtze River Delta, Pearl River Delta, Bohai Rim Area, and inland river areas, are less affected by boundary inflows from other

countries. In addition, our previous studies have demonstrated that this approach remains acceptable for regional simulations in China (Lv et al., 2020; Luo et al., 2024).

Furthermore, as shown in Table S3, the simulated O₃ concentrations agree well with ground-based observations, which supports the reliability and acceptability of our model results despite this potential limitation.

Revisions in Main Text:

2.4 Limitations

Anthropogenic emissions from other countries within the modeling domain were held fixed at 2010 levels, and open burning emissions were fixed at 2015 levels throughout the simulation period (2016–2020). Although this assumption simplifies the modeling framework and is unlikely to significantly alter the relative changes in shipping-related O₃ assessed in this work, it may still introduce some degree of uncertainty, particularly in regions where long-range transport or fire-related emissions could have contributed more dynamically during specific years. Future studies could benefit from incorporating temporally varying background emissions to further reduce potential uncertainties and improve the representation of external influences.

Comment 2

Page 3, lines 1-4. It should be mentioned that there are also effects of titration of ozone due to ship emissions especially at local scale, a few kilometres, that could complicate both simulation and data interpretation see Merico et al (Atmospheric Environment 139, 2016, 1-10).

Response:

Thanks for your comment. we have now added a discussion of this effect in the Introduction.

Revisions in Main Text:

1 Introduction

Additionally, the titration of O₃ by NO from shipping emissions, particularly within a few kilometers of ship tracks, can further complicate the simulation and interpretation of O₃

concentrations at the local scale (Merico et al., 2016).

Comment 3

Page 3, line 6. Is this a sufficient resolution to investigate local processes leading to ozone formation? Generally, modelling of these processes is done using a much more refined scale.

Response:

Thanks for your question.

We agree that a finer spatial resolution is generally more appropriate for capturing local-scale ozone formation processes. However, our objective in this study is to assess the regional and interannual impacts of shipping emissions on ozone pollution at the national scale, rather than focusing on local photochemical processes at the urban or neighborhood level.

Therefore, the selected resolution of $36 \text{ km} \times 36 \text{ km}$ represents a practical compromise between spatial detail and computational feasibility, especially considering the need to simulate multi-year scenarios (2016–2020) across the entire Chinese domain. This spatial resolution is also consistent with a series of studies by Geng et al. (as shown in the table below), who have extensively investigated ozone pollution and its driving mechanisms in China using similar model setups. We have added a statement in the Methods section.

Reference	Model/Spatial resolution
Drivers of Increasing Ozone during the Two Phases of Clean Air Actions in China 2013–2020	WRF-CMAQ/36 km
Evaluating the spatiotemporal ozone characteristics with high-resolution predictions in mainland China, 2013–2019	WRF-CMAQ/36 km
Estimating Spatiotemporal Variation in Ambient Ozone Exposure during 2013–2017 Using a Data-Fusion Mode	WRF-CMAQ/36 km

Additionally, the spatial resolution of the ship emission inventory we constructed is 0.05° , the land-based anthropogenic emission inventory from MEIC has a spatial resolution of 0.25° . Allocating land-based anthropogenic emissions to a much finer grid could significantly

increase the uncertainty of the simulation.

Revisions in Main Text:

2.4 Limitations

In this study, the spatial resolution of $36 \text{ km} \times 36 \text{ km}$ may not fully capture the fine-scale spatial heterogeneity of O_3 concentrations, particularly in coastal urban areas where emissions and photochemical reactions exhibit strong spatial variability. This resolution is relatively coarse for accurately representing O_3 exceedances and local photochemical processes, which often occur at much finer spatial scales. Consequently, localized O_3 peaks and gradients may be underestimated or smoothed in the model outputs. Despite this limitation, the selected resolution represents a practical compromise that enables multi-year simulations across the national domain.

Comment 4

Page 3, lines 31-32. What is Nm, nautical miles? Better to write it explicitly being not a SI unit.

Response:

Done.

Revisions in Main Text:

2.1 Shipping emissions

Here, emissions beyond 200 nautical miles from the Chinese mainland's territorial sea baseline were excluded from the domain by applying GIS-based spatial processing to the global shipping emission inventory, and only the annual shipping emissions from 2016 to 2020 within 200 nautical miles were used in the CMAQ-ISAM simulation.

Comment 5

The emissions used here, include the changes due to the implementation of IMO2020? It should be mentioned if it is expected an impact of this regulation on ozone formation due to shipping.

Response:

Thanks for your questions. The shipping emissions used in this study do account for the implementation of the IMO 2020 regulation.

Regarding the potential impact of IMO 2020 on ozone formation, although the regulation directly targets SO₂ and PM emissions, its indirect effects on O₃ may arise from increased VOC emissions. This is because low-sulfur fuels are typically richer in short-chain hydrocarbon (Wu et al., 2020). We have added a clarification in the manuscript to acknowledge this potential effect, although a detailed quantification of IMO 2020 impacts on O₃ formation is beyond the scope of this study and would require dedicated scenario analysis.

Revisions in Main Text:

2.1 Shipping emissions

Additionally, following the implementation of the global sulfur cap (IMO, 2018), the shift to low-sulfur fuels, which are typically richer in short-chain hydrocarbons (Wu et al., 2020), has contributed to a rise in shipping VOC emissions.

3.1 Annual O₃ impact from shipping emissions

Figure 4 illustrates the interannual trend in shipping-related O₃ in key regions from 2016 to 2020. Nationwide, the shipping-related O₃ shows a slight upward trend, with an average annual growth rate of 1.7%, primarily observed in coastal regions. This trend aligns with the changes in shipping NO_x and VOC emissions, especially in 2020 when a 0.2-0.3 ppb rise in shipping-related O₃ was observed, partly attributable to the notable increase in VOC emissions following the implementation of the global sulfur cap.

Comment 6

Page 4, line 18. Field rather than filed. In addition, why to use a one-year meteorology instead of the specific meteorology of each year? I believe that meteorological parameters have a strong influence on ozone formation and this is also what is mentioned in the conclusions..

Response:

Thanks for your comments. We have revised the typo error.

In this study, we primarily delve into the historical perspective of how anthropogenic emission changes impact shipping-related O₃. Consequently, we fixed the meteorological conditions to exclude their effects. We have now explained the reason for “fixing meteorological conditions” in the **2.2 Air quality model**.

Moreover, the impact of meteorological conditions should be insignificant. According to the National Climate Data Center (NCDC, <ftp://ftp.ncdc.noaa.gov/pub/data/noaa/>), the meteorological data (e.g., air temperature, relative humidity, monsoon) for the study area from 2016 to 2020 remained relatively stable (as shown in the Figure below). Additionally, based on the “China Climate Bulletin for the Year 2018”, the climate conditions in China for the year 2018 were overall normal, with few extreme weather events, making it a representative meteorological year. Therefore, we fixed the annual meteorological conditions in the year 2018. Furthermore, although there may have been some extreme weather events during that year, our focus on interannual PM_{2.5} variation minimizes the impact of these events.

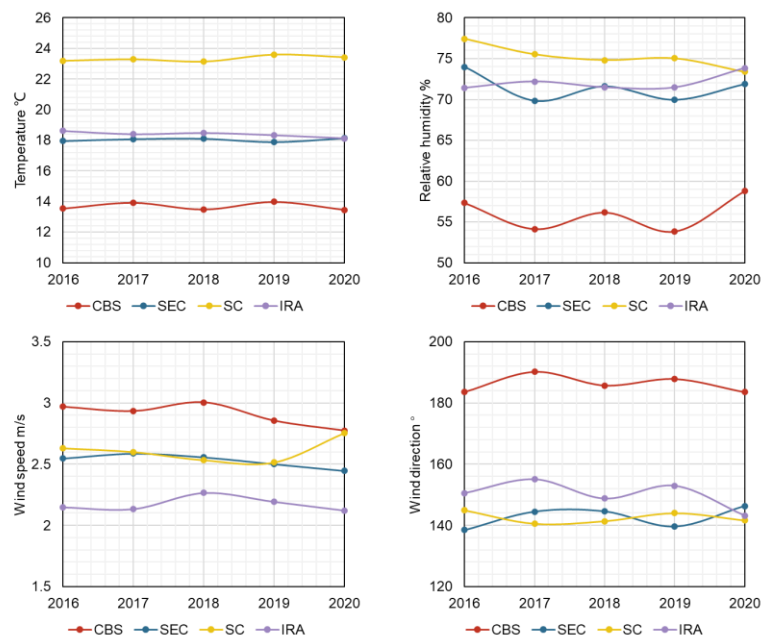


Figure The meteorological conditions for CBS, SEC, SC and IRD for 2016 to 2020

Revisions in Main Text:

2.2 Air quality model

Here, we primarily focused on examining the impact of anthropogenic emission changes on

shipping-related O₃ from a historical perspective. To eliminate the impact of interannual meteorological variability, we used meteorological field of 2018 (Zhao et al., 2022), which simulated by WRF and identified as a typical meteorological year due to its relatively stable climate conditions, to drive the CMAQ simulations for the period 2016-2020.

Comment 7

Page 7, lines 25-26. This sentence seems to say that shipping is not relevant for ozone formation and it is opposite to what is said in conclusions.

Response:

Thank you for pointing this out. We agree that the original sentence could be misinterpreted as suggesting that shipping emissions are not relevant to O₃ formation, which is not our intended meaning. Our point was that O₃ responds to precursor changes in a nonlinear variable manner, and the shipping-related O₃ increases are not directly proportional to the rise in shipping NO_x and VOC emissions. We have revised the sentence to clarify this and avoid confusion with the conclusion section.

Revisions in Main Text:

3.1 Annual O₃ impact from shipping emissions

This is because the formation of O₃ depends on photochemical reactions involving NO_x and VOC under solar radiation, and is influenced not only by the level of shipping emissions but also by land-based anthropogenic emissions, meteorological conditions, and long-range transport (Ye et al., 2023). Therefore, changes in shipping-related O₃ do not scale linearly with the changes in shipping NO_x and VOC emissions.

Comment 8

Figure 1. What is the cause of the increment of emission in 2020? Fig. S2 does not show a significant increase of cargo throughput. Could it be simply related to the use of a different emission database?

Response:

We appreciate the reviewer's comment. The emissions in 2020 were estimated using a consistent emission database and methodology across all years, ensuring comparability. While Figure S2 shows that cargo throughput did not increase substantially in 2020, the emission increment is likely driven by a combination of factors beyond throughput alone. These include changes in vessel operating conditions (e.g., increased idling time), variations in ship traffic patterns, and potentially longer operating durations of high-emitting vessels. We presented cargo throughput as a straightforward proxy, but acknowledge that it may not fully capture the complex dynamics influencing emissions. A more in-depth investigation would be needed to disentangle the contributing factors, which is beyond the scope of this study. Nonetheless, we have added a brief explanation of this complexity in the revised manuscript to provide additional context.

Revisions in Main Text:

2.1 Shipping emissions

It is worth noting that changes in vessel operating conditions, such as idling time and engine load, also influenced emissions.

Comment 9

Page 14, line 4 there is an “s” that should be eliminated..

Response:

Thank you for your careful reading. We carefully checked the sentence on Page 14, Line 4, but we were unable to identify an extra or incorrect use of “s” in that line.

References

- IMO, 2018. Resolution MEPC.305(73), Amendments to the Annex of the Protocol of 1997 to Amend the International Convention for the Prevention of Pollution from Ships.
- Luo, Z., Lv, Z., Zhao, J., Sun, H., He, T., Yi, W., Zhang, Z., He, K., Liu, H., 2024. Shipping-related pollution decreased but mortality increased in Chinese port cities. *Nature Cities* 1, 295-304.
- Lv, Z., Wang, X., Deng, F., Ying, Q., Archibald, A.T., Jones, R.L., Ding, Y., Cheng, Y., Fu, M., Liu, Y., 2020. Source–receptor relationship revealed by the halted traffic and aggravated haze in Beijing during the COVID-19 lockdown. *Environmental science & technology* 54, 15660-15670.

Merico, E., Donateo, A., Gambaro, A., Cesari, D., Gregoris, E., Barbaro, E., Dinoi, A., Giovanelli, G., Masieri, S., Contini, D., 2016. Influence of in-port ships emissions to gaseous atmospheric pollutants and to particulate matter of different sizes in a Mediterranean harbour in Italy. *Atmospheric Environment* 139, 1-10.