Manuscript id: acp-2022-394

Manuscript title: Contribution of marine biological emissions to gaseous methylamines in the atmosphere: an emission inventory based on satellite data

Responses to Reviewers' comments:

We thank the Reviewer for the constructive suggestions and helpful comments. We provide below itemized responses to each of the Reviewer's comments. The comments are given in bold while responses are in normal font. Changes made to the manuscript are shown in blue.

Reviewer #2:

Overall impression:

The study aims to estimate the gas phase concentration of three different amines using WRF-Chem. In addition to previous similar attempts using anthropogenic amine emissions only, the authors apply an online marine biogenic amine emission scheme. This new method seems sound and valid. However, the manuscript lacks of a thorough validation. (Some) Measurement results of different studies are presented in a table but never discussed in context with the simulated concentrations. Furthermore, the conclusions and discussion is not clearly formulated. For example, it is often unclear what type of aggregation / averages are referred to when presenting relative differences. This made it hard to follow the discussion and the results and likely lead to misunderstandings. Finally, potential shortcomings of the current method (e.g. loss to atmospheric aerosols is not considered) is not discussed anywhere in the paper.

Despite the fact that the manuscript could be improved in terms of language and clearer formulations at many points, the manuscript also needs major revision in the interpretation of results, discussion and conclusion. Since measured concentrations were presented, an evaluation section that compares these measurements, in particular the ones of the same year, with the modelled concentrations.

1) l. 138-139: Why is loss to aerosols not considered? Since it was correctly described in the introduction that amines play a role in aerosol formation, there should be a considerable loss to aerosols. This should be discussed here and in the conclusions.

Response:

We agree with the reviewer's opinion that absorption by aerosols is an important sink of amines. However, in this study, we used the aerosol scheme of CAM_MAM3_AQ which is unable to include the loss of amines to aerosols at present. There are some studies (e.g., Yu and Luo, 2014 and Mao et al., 2018) have considered the aerosol adsorption of amine simply by changing the uptake coefficient (γ) of amines to aerosol (instead of considering new particle formation involving amines) using different aerosol schemes (WRF-Chem with CB05 scheme by Mao et al. (2018) and GEOS-Chem v8.3.2 with an advanced particle microphysics (APM) model by Yu and Luo(2014)). However, with the CAM_MAM3_AQ employed in this study, more complex parameters are needed to consider the loss of amines to aerosol instead of simply including uptake coefficient. In this study, we used the aerosol scheme of CAM_MAM3_AQ because it is possible to new particle formation with amines (although not done yet in this study).

At present, only three species, namely H_2SO_4, NH_3 and MSA, are taken into consideration in CAM_MAM3_AQ. The uptake rate (uptkrate) of the latter two by aerosol is obtained by multiplying the uptake rate of H_2SO_4 into aerosol by a fixed value (uptkrate(NH_3)= uptkrate (H_2SO_4)×2.08, uptkrate(MSA)=uptake(H_2SO_4)×1.28). The uptake rate of H_2SO_4 is related to the particle size range, the concentration distribution of different particle sizes, and the diffusivity of H_2SO_4 gas molecules. At present, there is not enough literature to obtain accurate parameters related to different amines and H_2SO_4 in particles with different particle sizes. We plan to establish a new particle nucleation mechanism involving amines and embed it in the model based on laboratory simulation in a subsequent study. However, the current study did not consider the influence of aerosol absorption on amines. We have added relevant statements in the manuscript to explain why we did not consider aerosol absorption.

Lines 164-170: "Although absorption by aerosols is an important sink of amines, the current study did not consider the influence of aerosol absorption on amines. At present, only three species, namely H_2SO_4, NH_3 and MSA, are taken into consideration in CAM_MAM3_AQ. The uptake rate (uptkrate) of the latter two by aerosol is obtained by multiplying the uptake rate of H_2SO_4 into aerosol by a fixed value (uptkrate(NH_3) = uptkrate(H_2SO_4)×2.08, uptkrate(MSA)=uptake(H_2SO_4)×1.28). The uptake rate of H_2SO_4 is related to the particle size range, the concentration distribution of different particle sizes, and the diffusivity of H_2SO_4 gas molecules. At present, there is not enough literature to obtain accurate parameters related to different amines and H_2SO_4 in particles with different particle sizes."

2) l. 145-146: It is stated that 2015 was chosen since it had ‚more consecutive days with field observations', however, these field observations were not used in the manuscript for validation purposes. Furthermore, the authors cite e.g. Zheng et al., 2015, in this context, who performed measurements in 2012, so this study probably does not help to explain why 2015 was chosen.

Response:

Thanks for the comment. We are aware of the flaws in the setting of simulation periods in the manuscript. Therefore, we abandoned the discussion on seasonal variation and re-selected the simulation period for the following reasons:

① Although January, April, July and October were selected as representative months of winter, spring, summer and autumn for China (Cai et al., 2017) in the original manuscript, the actual simulation period was short and could not adequately represent the changing characteristics of a season.

② Insufficient observational data are available to evaluate the model's performance of the simulation of amines in different seasons. It is therefore doubtful whether the model's results in January, April and October are representative of the actual seasonal variation of atmospheric amines. Therefore, after the acquisition of new continuous observations, we believe that the simulation period should be re-selected based on the observations that can be used for verification.

③ In the original manuscript, the expression of seasonal variation of the amines simulation results is mainly related to the wind direction, and there are relatively detailed expressions only for July when the wind blows from the sea to the land, while the results of the other three simulation periods when the prevailing wind blows from the land to the sea are less expressed. In addition, the newly selected prevailing winds for December 2019 also blow from land to sea, so reselecting the simulation period does not affect the manuscript's main conclusions.

④ In addition, the new observational data are from marine sites, and hence, we can find out how the simulation improves for different types of sites after the addition of MBE.

The simulation period was reset according to the observation data obtained from the literature research. The continuous observation data of Shanghai urban site and Yellow and Bohai Seas marine site were selected for the verification of the simulation results. Thus, the effect of the amine emission inventory established in this study on the simulation effect of different types of sites was quantified. Because we re-selected the simulation period in the process of revising the manuscript and abandoned the expression of seasonal changes in the original manuscript, the description of seasonal changes in the applied satellite data was deleted.

Lines 172-177: " In this study, continuous observation data from Yao et al. (2016) collected in Shanghai urban site on 25-31 July 2015 (MMA, DMA, TMA were observed), and Chen et al. (2021) collected in the Yellow and Bohai Seas during 9–22 December 2019 (DMA, TMA were observed) were used for verification. Therefore, we simulated the amines concentration in July 2015 (2015.7.22 00:00:00 to 2015.7.31 18:00:00), and December 2019 (2019.12.6 00:00-2019.12.22 18:00) to explore the changes at urban and marine sites."

Lines 826-828: "

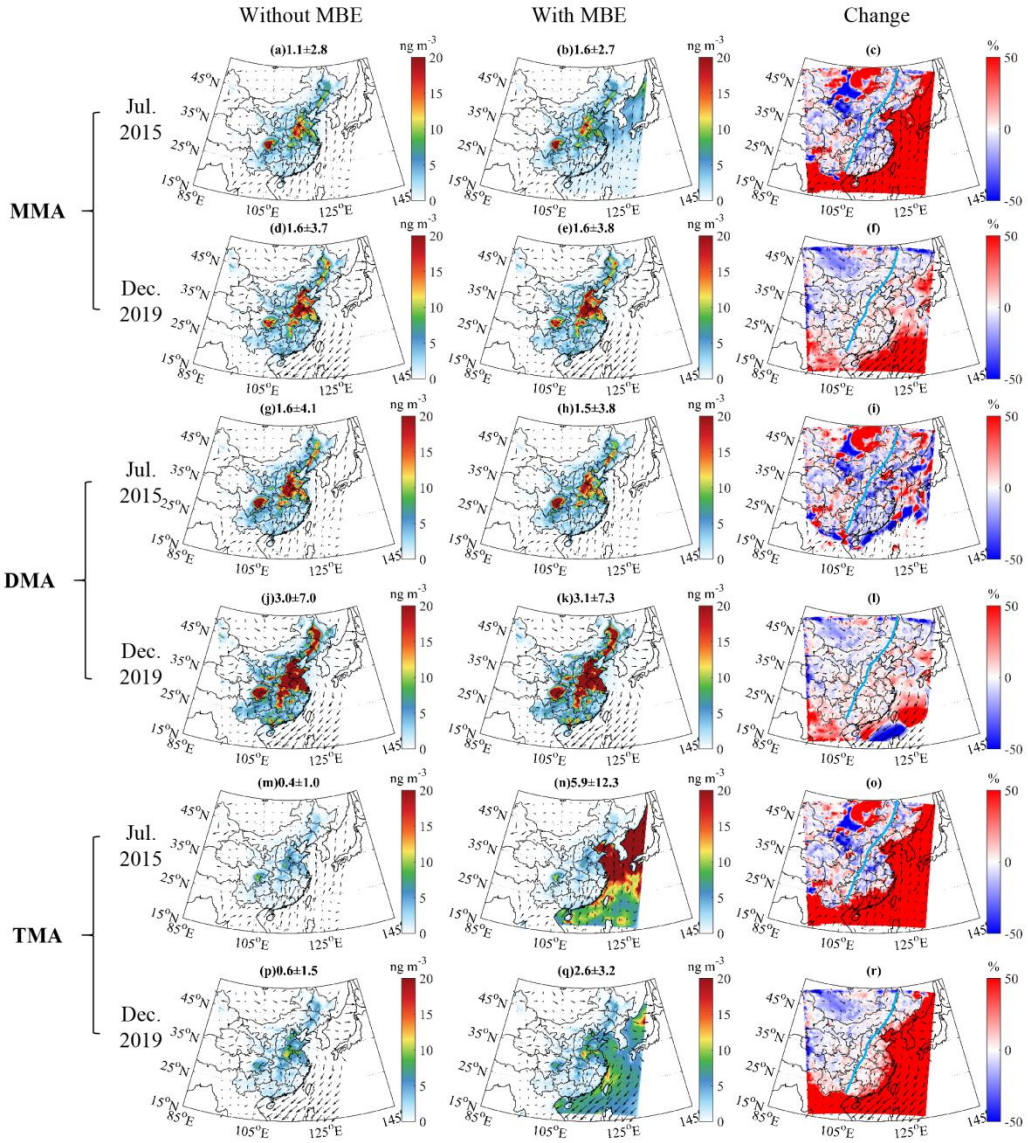


Figure 8: Spatial and temporal distribution of amines simulated mass concentration. (a-f): MMA. (g-l): DMA. (m-r): TMA. The solid blue line represents the boundary between the second and third steps of the Chinese terrain."

3) l. 148-149: The mentioned verification discussion is missing in the manuscript. l. 349-350: Weren't the simulation periods chosen to compare with observations? What were the available observations in the simulated time periods of 2015? Above it was mentioned, that daily measurements are available in certain periods. These periods were simulated. So why don't the authors take the grid cell where the measurement took place and produce a daily mean from the simulation and compare this to the observation?

Response:

We thank the reviewer for the constructive suggestion. We have added the discussion accordingly.

Lines:396-406: "The contribution of MBE to TMA improved the regional simulation value. Table 6 shows the simulation results of this study and the observation and simulation results

of other literature. Before adding MBE, the model significantly underestimated the concentration of amines in urban and marine sites. The addition of MBE resulted in a significant increase in the amines concentration simulated for marine sites (Fig. 7. 1116.7% for TMA and 9.0% for DMA.), effectively compensating for the model's underestimation. In addition, it is worth noting that the TMA concentration observed by Gao et al. (2021) and Chen et al. (2021, 2022) is significantly higher than that in other literatures. This may be because TMA is mainly affected by MBE, and hence, concentrations of TMA observed by Gao et al. (2021) and Chen et al. (2021, 2022) at marine sites are much higher than those observed at terrestrial sites. However, the simulated values at urban sites did not change significantly, indicating that the urban site is less affected by MBE in the simulation periods. It is found that the AE inventory established based on MEIC in this study has a low resolution, and some strong emission sources are missing in the Yangtze River Delta where the urban site are located, which leads to the underestimation of the amines concentration at the urban site."

Lines 824-825: "

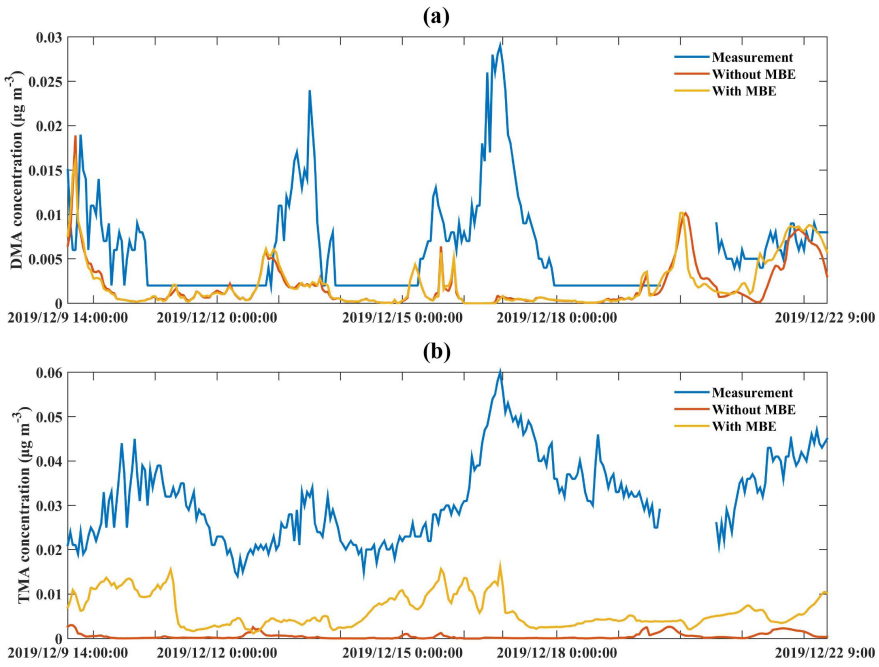


Figure 7: Comparison of observed and simulated amines concentrations at marine site (a) DMA, (b) TMA."

4) l. 351-352: Since the range of observed concentrations spans ~an order of magnitude for each of the three investigated amines, it's hard to say that simulation results are 'close'. What you could generally infer is that simulation and available observations agree in terms of order of magnitude. The increase due to MBE does not seem to be very strong seen the numbers in Table 6 to justify the statement 'inclusion of marine biogenic emissions can make up for the shortcomings of the previous models' underestimation of amines'

Response:

Thanks for the suggestion. In order to make up for the inadequacy of the argument mentioned by the reviewer, we have added in the manuscript a discussion of the simulation of urban site and marine site after the addition of MBE. At the same time, the error source of simulation is analyzed.

Lines:396-406: "The contribution of MBE to TMA improved the regional simulation value. Table 6 shows the simulation results of this study and the observation and simulation results of other literature. Before adding MBE, the model significantly underestimated the concentration of amines in urban and marine sites. The addition of MBE resulted in a significant increase in the amines concentration simulated for marine sites (Fig. 7. 1116.7% for TMA and 9.0% for DMA.), effectively compensating for the model's underestimation. In addition, it is worth noting that the TMA concentration observed by Gao et al. (2021) and Chen et al. (2021, 2022) is significantly higher than that in other literatures. This may be because TMA is mainly affected by MBE, and hence, concentrations of TMA observed by Gao et al. (2021) and Chen et al. (2021, 2022) at marine sites are much higher than those observed at terrestrial sites. However, the simulated values at urban sites did not change significantly, indicating that the urban site is less affected by MBE in the simulation periods. It is found that the AE inventory established based on MEIC in this study has a low resolution, and some strong emission sources are missing in the Yangtze River Delta where the urban site are located, which leads to the underestimation of the amines concentration at the urban site."

Lines 824-825: "

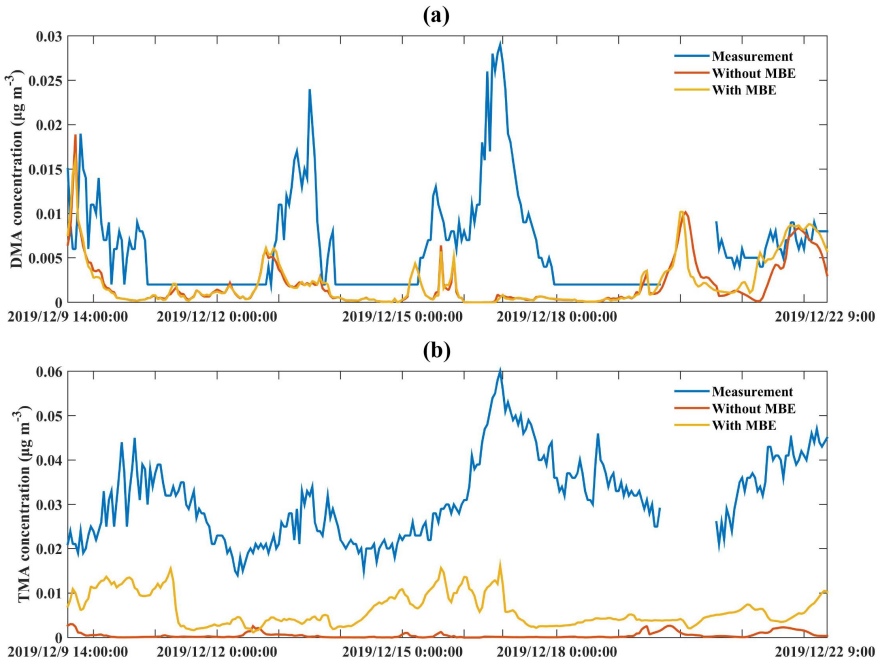


Figure 7: Comparison of observed and simulated amines concentrations at marine site (a) DMA, (b) TMA."

5) l. 355: The mentioned verification part is missing. In the text, there are no comparisons between measured and simulated values and no discussion of the evaluation of the simulated values against the observations shown in Table 6.

Response:

We have now added the discussion of validation in the manuscript.

Lines:396-406: "The contribution of MBE to TMA improved the regional simulation value. Table 6 shows the simulation results of this study and the observation and simulation results of other literature. Before adding MBE, the model significantly underestimated the concentration of amines in urban and marine sites. The addition of MBE resulted in a significant increase in the amines concentration simulated for marine sites (Fig. 7. 1116.7% for TMA and 9.0% for DMA.), effectively compensating for the model's underestimation. In addition, it is worth noting that the TMA concentration observed by Gao et al. (2021) and Chen et al. (2021, 2022) is significantly higher than that in other literatures. This may be because TMA is mainly affected by MBE, and hence, concentrations of TMA observed by Gao et al. (2021) and Chen et al. (2021, 2022) at marine sites are much higher than those observed at terrestrial sites. However, the simulated values at urban sites did not change significantly, indicating that the urban site is less affected by MBE in the simulation periods. It is found that the AE inventory established based on MEIC in this study has a low resolution, and some strong emission sources are missing in the Yangtze River Delta where the urban site are located, which leads to the underestimation of the amines concentration at the urban site."

Lines 824-825: "

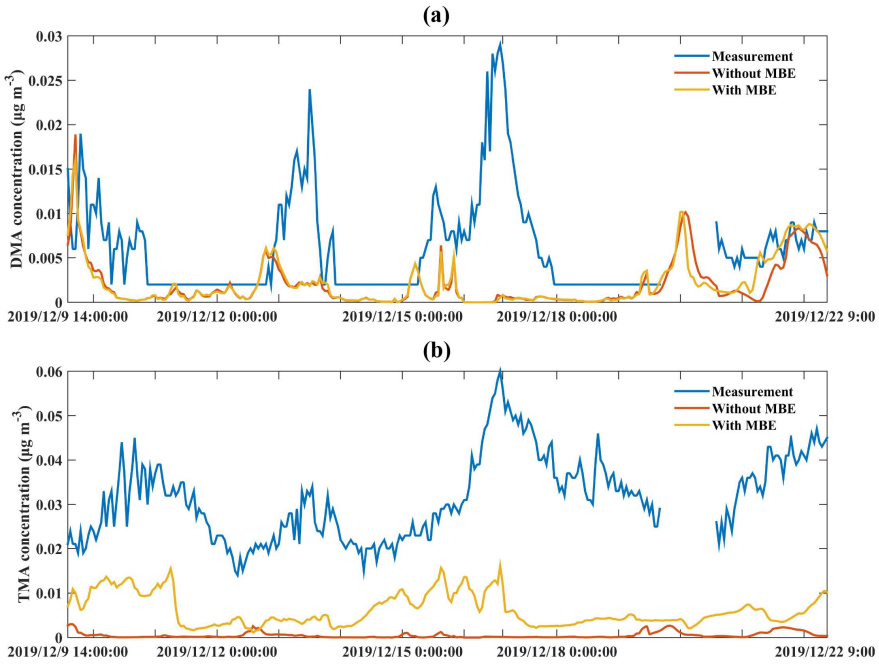


Figure 7: Comparison of observed and simulated amines concentrations at marine site (a) DMA, (b) TMA."

6) l. 364-365:

①Which region correspond these numbers to? I get different relative changes when using the numbers given in Table 6. Later I see, that these numbers seem to refer to only regions with an increase shown in Table 7, right? Why do the authors present these numbers and not the domain average in the text (MA in Table 7 if I understood correctly)? In any case, it need to be made much clearer in the whole discussion sections what averages the given numbers in the text refer to.

②At present it is impossible to follow the discussion of results in a fair amount of time.Conclusion section: It has to be stated that the drawn conclusions are only valid for the given months in the year 2015. What is with potential limitations since loss to atmospheric aerosol is not considered? This should be discussed. Overall none of the results of the sensitivity simulations in sections 3.3 is summarized in the conclusion. Why not? And a comparison to the available measurements is missing.

Response:

Thanks for the reviewer's comments.

①We are aware that the table here contains an incorrect number. At the same time, in the process of revising the manuscript, we thought that the content of the table mentioned was too little, and therefore, we deleted the table. In addition, we have explained other mean ranges mentioned in the manuscript.

②We have rewritten the Conclusions section to supplement the application scope of the conclusions, the influence of aerosol sinks on the simulation results, the comparison of simulation results with observations, and the summary of Section 3.3.

Lines 536-576: “The performance of the model for the numerical simulation of amines needs the support of accurate amines emission inventory. However, limited by high observation difficulty and complex environmental factors, the established inventory of amines emissions is still not comprehensive enough. For example, the gridded inventory of emissions from animal husbandry and marine organisms is still scarce. Therefore, in order to improve the model simulation performance of ambient gaseous amines (MMA, DMA, and TMA), we established the AE inventory based on the ratio method, and established gridded MBE inventory using easily available satellite data such as Chla, SST, SSS and NH_3.

From the calculation, we realize that the ocean can be either a source or a sink for MMA; for DMA, the ocean always appears as a sink; for TMA, the ocean always appears as the source. However, if the ratio method is adopted, whether the ocean is the source of amines emission depends entirely on whether ammonia can be discharged from the ocean. This "one-size-fits all" method is not consistent with the actual situation, and further affects the simulation results of subsequent models. The emission scheme calculated from satellite data can better reflect the exchange direction of amines between ocean and atmosphere and reflect the emission characteristics of different amines.

Due to the contribution of MBE, the ambient concentration of TMA of coastal areas increased multiple times, and the concentration of amines above the coastal area also increased significantly. I) In July 2015 and December 2019, TMA increased by 43917.0%, and 804.0%, respectively. The average increase rates of TMA in the two periods even reached 50% over

567 km and 378 km from the coastline. II) The increase of MMA concentration is less than that of TMA, with an average of 2635.4% and 0.37% in July 2015 and December 2019, respectively. III) Moreover, due to the obvious increase of the other two amines, ·OH decreased and the consumption of DMA decreased correspondingly, thus leading to a small change in the DMA concentration (-3.9% in July 2015, and 1.1% in December 2019). The addition of MBE also resulted in a significant increase in the amines concentration simulated for the selected marine sites (1116.7% for TMA and 9.0% for DMA.), effectively compensating for the model's underestimation. However, the simulated values at the urban site did not change significantly, indicating that the selected urban site is less affected by the MBE in the simulation periods. It is found that the AE inventory established based on MEIC in this study has a low resolution, and some strong emission sources are missing in the Yangtze River Delta where the urban site is located, which leads to an underestimation of amines concentration at the urban site.

WS, Chla, and $[C^{+}_{(s)tot}]$ were found to be the dominant factors affecting MBE fluxes for all of the above amines. WS is directly used in the calculation of the air–ocean transfer velocity (K_g), and the influence of MBE is conditioned by the monsoon and the topography of China. The prevailing southwest monsoon in July can thrust the marine air mass into the third step area (the continental shelf), and its influence area is wider due to the lack of obstacles in the north-south direction. The special geographical characteristics of some areas, e.g., Hainan Island, Fujian and Guangdong provinces etc., make them more prone to the influence of the strong ocean air mass. Chla indirectly influences the calculation results of exchange flux by affecting the calculation of pH. While increasing $[C^{+}_{(s)tot}]$ means increasing the current dissolved amines in seawater ($[C_{(s)}]$), the ocean tends to behave as a source. In addition, the emission flux and spatial distribution of AE, and wet deposition also affect the simulation of amines.

However, it should be noted that due to the limitation of the algorithm (e.g., Eq. (7)), the MBE established in this study is only applicable to the North Pacific region. In addition, this study compares the model results with the observed values of the two types of sites, and the simulation period is short; therefore, the subsequent simulation results and observation data should be longer to explore the application effect of this method in different seasons. In addition, the influence of aerosol and ocean as sinks of amines concentration was not considered in the model in this study, which may lead to an overestimation of the contribution of MBE to amines concentration in the model results. Therefore, further studies are needed to establish a more accurate numerical simulation for the amines."

7) l. 484-486: The authors should summarize what they actually did. This numerical modelling to simulate gas phase amine concentration. In addition to anthropogenic emission MBE of amines was treated in an online manner.

Response:

Thanks for the reviewer's suggestion. We have rewritten the conclusion and abstract of the manuscript.

Lines 17-40: "Methylamines can readily react with acidic gases in the atmosphere, which consequently has an important impact on the atmospheric environment. It is difficult to measure amines in field studies due to their high reactivity, and therefore, numerical

modelling is an effective tool to study ambient amines. In order to improve the model simulation performance of ambient gaseous amines (monomethylamine (MMA), dimethylamines (DMA), and trimethylamines (TMA)), we established the anthropogenic emissions (AE) inventory based on the ratio method, and established gridded MBE inventory using easily available satellite data (Sea Surface Temperature (SST), Chlorophyll-a (Chla), Sea Surface Salinity (SSS), NH_3 column concentration (NH_3)), and model simulation data (Wind Speed, WS) to establish a more reasonable MBE inventory of amines. From the calculation, we realized that the ocean can be either a source or a sink for MMA; for DMA, the ocean always appears as a sink; for TMA, the ocean always appears as the source. Due to the contribution of MBE, the ambient concentration of TMA of coastal areas increased multiple times, and the concentration of amines above the coastal area also increased significantly. In July 2015 and December 2019, TMA increased by 43917.0%, and 804.0%, respectively. The average increase rates of TMA in the two periods even reached 50% over 567 km and 378 km from the coastline. The increase of MMA concentration is less than that of TMA, with an average of 2635.4% and 0.37% in July 2015 and December 2019, respectively. Moreover, due to the obvious increase of the other two amines, ·OH decreased and the consumption of DMA decreased correspondingly, thus leading to a small change in the DMA concentration (-3.9% in July 2015, and 1.1% in December 2019). The addition of MBE also resulted in a significant increase in the amines concentration simulated for the selected marine sites (1116.7% for TMA and 9.0% for DMA.), effectively compensating for the model's underestimation.

WS, Chla, and $[C^{+}_{(s)tot}]$ were found to be the dominant factors affecting MBE fluxes for all of the above amines. WS is directly used in the calculation of the air–ocean transfer velocity (K_g), and the influence of MBE is conditioned by the monsoon and the topography of China. Chla indirectly influences the calculation results of exchange flux by affecting the calculation of pH. While increasing $[C^{+}_{(s)tot}]$ means increasing the current dissolved amines in seawater ($[C_{(s)}]$), the ocean tends to behave as a source. In addition, the emission flux and spatial distribution of AE, and wet deposition also affect the simulation of amines."

Lines 536-576: "The performance of the model for the numerical simulation of amines needs the support of accurate amines emission inventory. However, limited by high observation difficulty and complex environmental factors, the established inventory of amines emissions is still not comprehensive enough. For example, the gridded inventory of emissions from animal husbandry and marine organisms is still scarce. Therefore, in order to improve the model simulation performance of ambient gaseous amines (MMA, DMA, and TMA), we established the AE inventory based on the ratio method, and established gridded MBE inventory using easily available satellite data such as Chla, SST, SSS and NH_3.

From the calculation, we realize that the ocean can be either a source or a sink for MMA; for DMA, the ocean always appears as a sink; for TMA, the ocean always appears as the source. However, if the ratio method is adopted, whether the ocean is the source of amines emission depends entirely on whether ammonia can be discharged from the ocean. This "one-size-fits all" method is not consistent with the actual situation, and further affects the simulation results of subsequent models. The emission scheme calculated from satellite data can better reflect the exchange direction of amines between ocean and atmosphere and reflect the

emission characteristics of different amines.

Due to the contribution of MBE, the ambient concentration of TMA of coastal areas increased multiple times, and the concentration of amines above the coastal area also increased significantly. I) In July 2015 and December 2019, TMA increased by 43917.0%, and 804.0%, respectively. The average increase rates of TMA in the two periods even reached 50% over 567 km and 378 km from the coastline. II) The increase of MMA concentration is less than that of TMA, with an average of 2635.4% and 0.37% in July 2015 and December 2019, respectively. III) Moreover, due to the obvious increase of the other two amines, ·OH decreased and the consumption of DMA decreased correspondingly, thus leading to a small change in the DMA concentration (-3.9% in July 2015, and 1.1% in December 2019). The addition of MBE also resulted in a significant increase in the amines concentration simulated for the selected marine sites (1116.7% for TMA and 9.0% for DMA.), effectively compensating for the model's underestimation. However, the simulated values at the urban site did not change significantly, indicating that the selected urban site is less affected by the MBE in the simulation periods. It is found that the AE inventory established based on MEIC in this study has a low resolution, and some strong emission sources are missing in the Yangtze River Delta where the urban site is located, which leads to an underestimation of amines concentration at the urban site.

WS, Chla, and $[C^{+}_{(s)tot}]$ were found to be the dominant factors affecting MBE fluxes for all of the above amines. WS is directly used in the calculation of the air–ocean transfer velocity (K_g), and the influence of MBE is conditioned by the monsoon and the topography of China. The prevailing southwest monsoon in July can thrust the marine air mass into the third step area (the continental shelf), and its influence area is wider due to the lack of obstacles in the north-south direction. The special geographical characteristics of some areas, e.g., Hainan Island, Fujian and Guangdong provinces etc., make them more prone to the influence of the strong ocean air mass. Chla indirectly influences the calculation results of exchange flux by affecting the calculation of pH. While increasing $[C^{+}_{(s)tot}]$ means increasing the current dissolved amines in seawater ($[C_{(s)}]$), the ocean tends to behave as a source. In addition, the emission flux and spatial distribution of AE, and wet deposition also affect the simulation of amines.

However, it should be noted that due to the limitation of the algorithm (e.g., Eq. (7)), the MBE established in this study is only applicable to the North Pacific region. In addition, this study compares the model results with the observed values of the two types of sites, and the simulation period is short; therefore, the subsequent simulation results and observation data should be longer to explore the application effect of this method in different seasons. In addition, the influence of aerosol and ocean as sinks of amines concentration was not considered in the model in this study, which may lead to an overestimation of the contribution of MBE to amines concentration in the model results. Therefore, further studies are needed to establish a more accurate numerical simulation for the amines."

Minor comments

1) Please check the manuscript with a native English speaker if possible. At many instances, some formulations were misleading or seemed at least very unfamiliar.

Response:

We have checked the manuscript and modified accordingly.

2) l. 20: ‚has been insufficiently investigated in the current emission inventory.' What do the author's want to express? An emission inventory cannot investigate.

Response:

We have modified the inappropriate words here.

Lines 19-24: "In order to improve the model simulation performance of ambient gaseous amines (monomethylamine (MMA), dimethylamines (DMA), and trimethylamines (TMA)), we established the anthropogenic emissions (AE) inventory based on the ratio method, and established gridded MBE inventory using easily available satellite data (Sea Surface Temperature (SST), Chlorophyll-a (Chla), Sea Surface Salinity (SSS), NH_3 column concentration (NH_3)), and model simulation data (Wind Speed, WS) to establish a more reasonable MBE inventory of amines. "

3) l. 60-62: Stronger and more easily oxidized than what? Likely ammonia, then please state it like this.

Response:

We have revised the imprecise expression here.

Lines 68-72: "It is believed that amines have stronger reactivity and are more easily oxidized by oxidants ($\cdot NO_3$, $\cdot OH$ and O_3) than NH_3, are condensed into particulates and scavenged from the atmosphere by both wet and dry depositions (Carl and Crowley, 1998; Barsanti et al., 2009; Qiu et al., 2011; Qiu and Zhang, 2013; Yu and Luo, 2014; Yao et al., 2016; Mao et al., 2018; Waller et al., 2018)."

4) l. 63-65: Is the lower concentration of amines compared to ammonia really a result of faster loss processes only or at least partly due to the lower emission rates?

Response:

We have added necessary instructions to make the conclusion here more rigorous.

Line 72: "And the emission of amines is significantly lower than that of ammonia (Ge et al., 2011a), as a result..."

5) l. 76-80: It is referred to ‚increased concentrations', an ‚increase by 1-2 orders of magnitude' and a ‚significant increase', but it is not mentioned compared to what concentrations have increased?

Response:

We have added the objects of comparison here.

Line 87: "...than those at the semi-urban site..."

6) l. 89-90: Not sure if China's location alone can be called unique. Densely populated coastal areas can be found in many places in the world. The combination of location and pollution is rather what could be called unique.

Response:

We have modified the imprecise expression here.

Lines 99-100: “Due to its special geographical location and pollution composition, eastern China was chosen as the research area in this study.”

7) l. 95: ‚amines' role in atmospheric chemistry‘ sounds strange. Better: ‚the role of amines in …'?

Reponse:

We have corrected the grammatical errors here.

Lines 103-105: “Accurate emission inventories of amines can lead to better model simulations of its atmospheric levels and corresponding spatiotemporal variations, and it in turn can lead to a better understanding of the role of amines in atmospheric chemistry and aerosol formation.”

8) l. 101: Is ‚reasonable‘ the right word? ‚Complete‘ or ‚detailed‘ might fit better.

Response:

Thanks for the reviewer's suggestion, we have replaced the inappropriate words here.

Line 111: “..., which is the most detailed emission inventory of amines to date.”

9) l. 102-103: First, is the emission mass ratio in the Mao et al., 2018, AE emission inventory really ‚arbitrary'? Second, since this scheme is used as it is, and just an online MBE scheme was added, your study does not ‚overcome the arbitrariness' in the AE emission inventory. Please reformulate these sentences in a more clearer way.

Response:

Thanks for the reviewer's suggestion, we have rewritten the unclear expression here.

Lines 111-116: “However, as stated by Mao et al. (2018), there are still great uncertainties in determining the mass ratio of amine to ammonia emission (e.g., animal husbandry, etc. Chang et al., 2021). And the application scope is limited, for example, has not been applied to the establishment of MBE. The above factors affect the simulation performance of the model for amines. Therefore, we attempt to establish an emission inventory of amines including MMA, DMA, and TMA from MBE based on their mechanisms of production.”

10) l. 104: Multiple -> multiple

Response:

We have modified this word.

Lines 116-118: “Model simulated wind speed and multiple satellite datasets including Sea Surface Temperature (SST), Chlorophyll-a (Chla), Sea Surface Salinity (SSS), NH_3 column concentration (NH_3) were incorporated to calculate the emission fluxes of amines.”

11) l. 121: This is confusing here. Which is the ‚previous method' you are referring to?

Response:

We have provided relevant details.

Lines 134-135: "... (Myriokefalitakis et al., 2010; Yu and Luo, 2014; Bergman et al., 2015) ..."

12) l. 127-129: Is an ammonia / amine ratio, e.g. similar to traffic, also applied for ammonia emissions from ships? If not, why not?

Response:

Yes. We applied the amine/ammonia ratio of transportation emissions for ammonia emissions from ships.

13) l. 150: What do the authors mean with 'representative'? Do these months represent a specific season? If so, is this also the case for these months in the year 2015? As the authors state later, at least SST might be non-typical for parts of the 2015, which indicates it is not representative in general. So, please clarify what is meant with ‚representative' in the context of this paper. I doubt that 10 days of one month can be called ‚representative' for the same month in a climate view or in other years.

Response:

Thanks for the comment. We agree with the reviewer that the simulation periods set are not persuasively representative. Therefore, the discussion on seasonal variation was abandoned, and the simulation period was reset according to the observation data obtained from literature research. The continuous observation data of Shanghai urban site and Yellow and Bohai Seas marine site were selected for the verification of the simulation results. Thus, the effect of the amine emission inventory established in this study on the simulation for different types of sites was quantified.

Lines 172-177: "In this study, continuous observation data from Yao et al. (2016) collected in Shanghai urban site on 25-31 July 2015 (MMA, DMA, TMA were observed), and Chen et al. (2021) collected in the Yellow and Bohai Seas during 9–22 December 2019 (DMA, TMA were observed) were used for verification. Therefore, we simulated the amines concentration in July 2015 (2015.7.22 00:00:00 to 2015.7.31 18:00:00), and December 2019 (2019.12.6 00:00-2019.12.22 18:00) to explore the changes at urban and marine sites."

14) l. 154: ‚chemical boundary' -> ‚chemical boundary conditions'

Response:

We have modified the text here.

Lines 177-178: "In addition, the chemical boundary conditions used in this study was the CAM-CHEM results provided by NCAR (National Center for Atmospheric Research)."

15) l. 154-155: Did the authors run CAM-CHEM themselves to generate boundary conditions? This is what I understand from reading. However, the webpage given a few sentences later suggest that driving data for boundary conditions can be download. Please clarify.

Response:

We used the CAM-CHEM simulation data provided by NCAR as chemical boundary conditions. We also revised the manuscript where it was unclear.

Lines 177-178: "In addition, the chemical boundary conditions used in this study was the CAM-CHEM results provided by NCAR (National Center for Atmospheric Research)."

Lines 181-182: "
https://wiki.ucar.edu/display/camchem/CESM2.1%3ACAM-chem+as+Boundary+Conditions"

16) l. 168: Why the reference to these measurements of concentrations in air and in water by Gibb et al., 1999? How exactly were these used?

Response:

We have revised the use of relevant data and have revised the manuscript.

Line 192: "...method of Section 2.5.4..."

Lines 297-310: "

2.5.4 NH_3

Ammonia total columns retrieved from IASI measurements from the ANNI-NH3-v2.1R-I retrieval algorithm (https://iasi.aeris-data.fr/nh3/) was used to calculate gaseous amines concentration (e.g., $MMA_{(g)}$, $DMA_{(g)}$, $TMA_{(g)}$). The empirical formula established by Yu et al. (2019) was used to estimate the ground NH_3 concentration, as shown below.

$$[NH_3]_G = 0.3413 \times 10^{-15} \times [NH_3]_R \quad (12)$$

$[NH_3]_G$ represents ground concentration measurements for NH_3 (Fig. 2(d, i)); $[NH_3]_R$ represents NH_3 column data (units: molec cm^{-2}). In this study, the data of July 2015 and December 2016 (December 2019 data is incomplete) were adopted.

The estimates of MMA(g), DMA(g) and TMA(g) are based on the linear relationship between amine and ammonia established by Zheng et al. (2015) after field observations in the northern part of Nanjing from August to September 2012. The regression equation is shown below,

$$MMA_{(g)} = 0.85 \times NH_{3(g)} + 0.83 \quad (13)$$

$$DMA_{(g)} = 1.56 \times NH_{3(g)} + 1.28 \quad (14)$$

$$TMA_{(g)} = 0.37 \times NH_{3(g)} + 0.41 \quad (15)$$

where the unit of amines concentration is pptv, and the unit of NH_3 concentration is ppbv."

17) l. 188: solvated -> dissolved?

Response:

We have corrected the inappropriate word here.

Line 211: "$[C^+_{(s)}]$ is the concentration of corresponding dissolved cation..."

18) l. 189-190: ‚this study selected the average of the observed values of other sea areas (Table 3)'. I don't understand what the authors want to express here. Which numbers of

Table 3 are used in the emission algorithm and how exactly? The description of your method might need more detail or clarification.

Response:

The total dissolved concentration of analyte (nM, i.e., $NH_4^+{}_{(s)tot}$, $MMAH^+{}_{(s)tot}$, $DMAH^+{}_{(s)tot}$, $TMAH^+{}_{(s)tot}$), from Table 3 was used to calculate the concentration of unionized solute molecules ($C_{(s)}$). We have also added a more detailed description of the data application method.

Lines 213-218: " In this study, the mean value of all $[C_{(s)\ tot}{}^+]$ observations in the same quarter is used for the relevant calculation for the target month. If the observation time is not specified, the data is included in the calculation of the mean value; if the observation value is not specified (i.e., if only the variation range of observation value is given), the data is not included in the calculation. In the simulation period (July 2015, and December 2019), the $[MMAH^+{}_{(s)tot}]$ was 36.1 nM and 38.9 nM, respectively, $[DMAH^+{}_{(s)tot}]$ was 6.0 nM and 9.8 nM, and $[TMAH^+{}_{(s)tot}]$ was 6.8 nM and 7.6nM, respectively."

19) l. 198-199: Perhaps related to the previous comment… It would be helpful if the authors can give their calculated average values in Table 3. This helps the reader to much easier follow the method.

Response:

We have added the average values used in this study.

Lines 213-218: "In this study, the mean value of all $[C_{(s)\ tot}{}^+]$ observations in the same quarter is used for the relevant calculation for the target month. If the observation time is not specified, the data is included in the calculation of the mean value; if the observation value is not specified (i.e., if only the variation range of observation value is given), the data is not included in the calculation. In the simulation period (July 2015, and December 2019), the $[MMAH^+{}_{(s)tot}]$ was 36.1 nM and 38.9 nM, respectively, $[DMAH^+{}_{(s)tot}]$ was 6.0 nM and 9.8 nM, and $[TMAH^+{}_{(s)tot}]$ was 6.8 nM and 7.6nM, respectively."

20) l. 208: The unit is probably is 45 g kg-1. The term ‚within' suggest a range but only one number is given.

Response:

Thanks for the reviewer's reminding, we have modified the inappropriate words here. "45" here is a dimensionless parameter.

Lines 236-237: "...the calculation of pK_a can be obtained from the empirical equation provided by Khoo et al. (1977) and applicable when the water salinity is less than 45."

21) l. 226-228: Was this done by the authors? If yes, please describe in more detail how the SST data was combined with the other satellite data sets mentioned here. According to the description before, the NESDIS SST data is already on 5 km horizontal resolution.

Response:

The analysis is carried out by NESDIS. We have clarified this as follows:

Lines 258-260: “ In the analysis, NESDIS combines SST data from the US, Japanese and European Synchronous Infrared Imagers and Low Earth Orbit Infrared (US and Europe) to produce a set of high-resolution (5km) SST data products.”

22) l. 235: ‚SST presents the seasonality‘ sounds unfamiliar. Please check for proper English.

Response:

We have now re-selected the simulation period in the process of revising the manuscript and have deleted the description of SST seasonal changes.

23) l. 257: Obsolete "at the".

Response:

We have modified the sentence here.

Lines 281-283: “On the one hand, human activities discharge nutrients into the ocean from the estuaries of Liaohe River, the Yellow River, Huaihe River, the Yangtze River and the Pearl River.”

24) l. 275: ion intensity -> ionic strength?

Response:

We have modified the phrase (Lines 195, 236, 291).

25) section 2.5.4: Is time-resolved (e.g. hourly) wind speed utilized or monthly mean?

Response:

The wind speed used in the study is hourly. We have added relevant instructions to the manuscript.

Line 312: “...the hourly WS...”

26) l. 293: Meaning of FNL is missing.

Response:

We have added the meaning of FNL in the manuscript.

Line 313: “...FNL (Final Reanalysis Data; It is produced by NCEP and NCAR).”

27) l. 294-295: Well, there is considerable bias for RH. This might not be important for your study, but in any case should not be stated suggestion good agreement. Most importantly is that model and observations of wind speed agree reasonably well.

Response:

Thanks for the suggestion. We have modified the expression here.

Lines 314-316: “It can be seen from the results that the model can generally simulate the numerical values and variation trends of WS, temperature and relative humidity.”

28) l. 302: Are the numbers presented in Table 5 for one grid cell or domain mean? Or only over ocean?

Response:

The numbers presented in Table 5 are for the domain over ocean. We have modified the caption of Table 5 to make this clearer.

Lines 803-804: “

Table 5 MBE fluxes of the three types of amines over ocean in July 2015 and Deember 2019.

Site	Date	MMA (pmol m^{-2} s^{-1})	DMA (pmol m^{-2} s^{-1})	TMA (pmol m^{-2} s^{-1})	Ref.
Waters east of China	July 2015 December 2019	-0.81±0.90 -0.13±0.20	-1.9±1.7 -0.86±0.38	2.8±1.3 5.2±1.1	This study
Coastal Hawaii and Massachusetts	-	0.11—1.80	- 0.46— -0.49	0.20—3.20	Van Neste et al. (1987)
The island of Sao Vicente	November. 2013	-0.40—0.087	-2.17—1.9	-	Van Pinxteren et al. (2019)

”

29) l. 303-307: Why not also in table 5 change the signs, which is then consistent with the rest of the figures and the paper?

Reponse:

Thanks for the suggestion. We have modified the signs in Table 5.

Lines 803-804: “

Table 5 MBE fluxes of the three types of amines over ocean in July 2015 and Deember 2019.

Site	Date	MMA (pmol m^{-2} s^{-1})	DMA (pmol m^{-2} s^{-1})	TMA (pmol m^{-2} s^{-1})	Ref.
Waters east of China	July 2015 December 2019	-0.81±0.90 -0.13±0.20	-1.9±1.7 -0.86±0.38	2.8±1.3 5.2±1.1	This study
Coastal Hawaii and Massachusetts	-	0.11—1.80	- 0.46— -0.49	0.20—3.20	Van Neste et al. (1987)
The island of Sao Vicente	November. 2013	-0.40—0.087	-2.17—1.9	-	Van Pinxteren et al. (2019)

”

30) l. 308, Fig. 3: From table 5 I understand that the ocean is a source for MMA and TMA, but a sink for DMA. Why aren't therefore negative emission fluxes seen for DMA over ocean in Fig. 3?

Response:

Fig. 3 shows the spatial distribution of the emission flux in the emission files used by the model. However, if the emission flux is negative, the model cannot be applied. Therefore, the influence of ocean sinks on DMA is not considered in this study. We have added a clarification in this regard.

Lines 324-326: "However, if the emission flux is negative, the model cannot be applied. Therefore, the influence of ocean sinks on DMA and MMA is not considered in this study."

31) l. 309, Fig. 4: Is that domain mean?

Response:

Yes. We have changed the caption of Fig.4 to make it clearer.

Lines 814-816: "

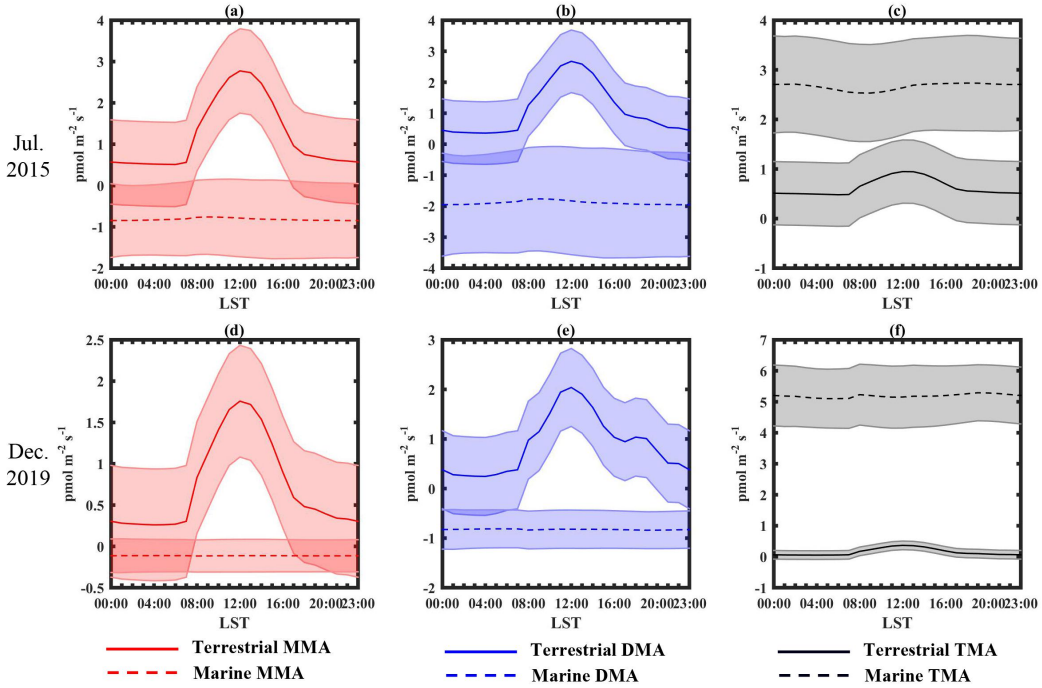


Figure 4: Diurnal variation of amines emission fluxes average. (a, d) MMA, (b, e) DMA. (c, f) TMA. The solid lines represent the terrestrial AE fluxes, the dotted lines represent the MBE fluxes. The LST means the Beijing time(UTC+08:00)."

32) l. 309: , MBE emission ' -> emission to much, since the E in MBE already refers to emission.

Response:

Since the content on seasonal variations was removed from the revised manuscript, the text mentioned by the reviewer has been deleted. We have now corrected similar errors in the manuscript.

33) l. 310, 311: TMA -> , MBE flux of TMA ' or similar.

Response:

We have revised the expression in the manuscript.

Lines 327-328: "As we can see, the MBE fluxes of TMA are quite substantial, which are significantly higher than terrestrial AE fluxes."

34) l. 309-314: Why no discussion of DMA emission flux, but only for MMA and TMA?

Response:

We have added the discussion of DMA to the manuscript.

Lines 330-333: “Compared with the other two amines, AE fluxes of DMA showed a "bimodal" diurnal variation due to a higher proportion of residential emission, with high values appearing around LST12:00 and LST20:00. Similarly, due to the small diurnal variation of marine environment, the diurnal variation of exchange flux is not significant.”

35) l. 315-335: Just as a suggestion: These paragraphs might be more useful directly after section 2.5, before section 3.1.

Response:

Thanks for the suggestion. However, we consider the emission flux of amines as the result of calculation of the original data. In addition, it leads to a discussion on influencing factors of emission flux estimation, which echoes the discussion on influencing factors of simulation results in Section 3.3. Therefore, the original structure of this manuscript is maintained.

36) l. 315, Fig. 5: Which values were used for the other three input variables, while only one is varied? I understand that the four plots do not show the contribution, but the variation of the emission flux due to the variation of one variable while the others were held constant. It is important for the inter pretation and the magnitude of the contribution to provide a clearer description here.

Please also clarify which area or aggregation Fig. 5 refers to. Is that temporal domain mean or ocean mean or one grid cell?

Response:

We have added the introduction of sensitivity experiment method here. In addition, we have changed the caption of Fig. 5 to clarify the meaning of the data shown.

Lines 334-336: “The influence of WS, Chla, SST, SSS, NH_3, and [$C^+_{(s)tot}$] on the exchange fluxes is shown in Fig. 5. The control variable method was used to obtain the influence of different factors on MBE fluxes by changing one of the variables and maintaining the mean value of the sea area for the other three variables.”

Lines 818-821: “

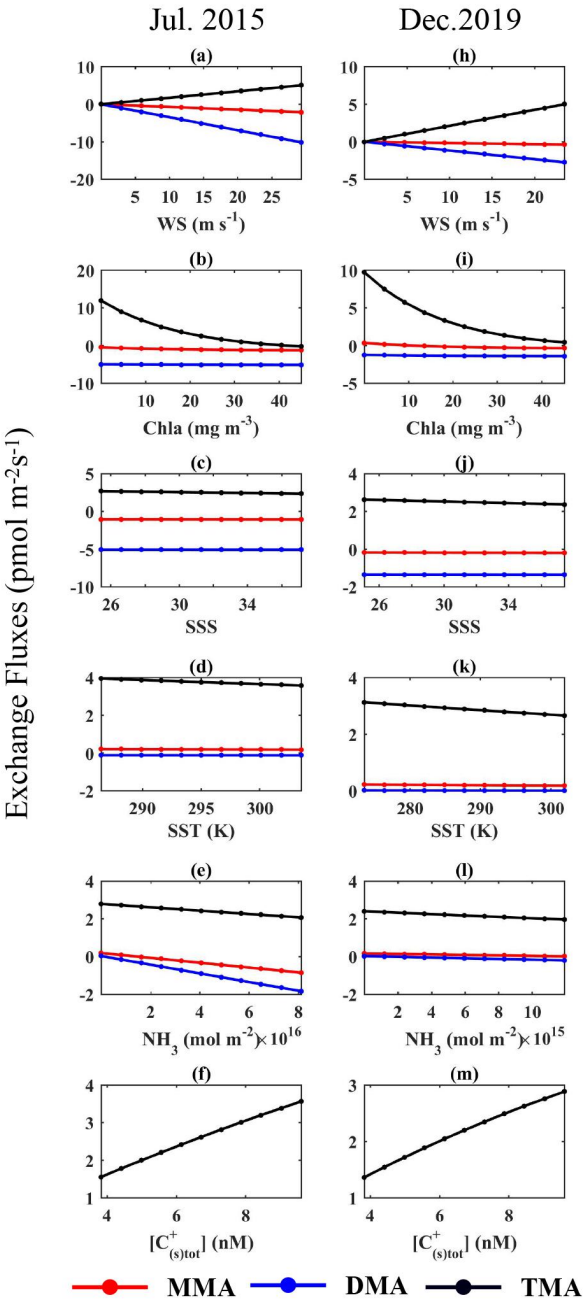


Figure 5: Influence of WS (a, h), Chla (b, i), SSS (c, j), SST (d, k), NH_3 (e, l), and $[C^{+}_{(s)tot}]$ (f, m) on the average of MBE. In order to ensure the simplicity of the figure, only the changes of TMA are plotted here, and the changes of the other two amines are consistent with TMA."

37) l. 316: , Kg, which is directly proportional to Kg' -> Probably the latter Kg is meant to be something else.

Response:

We have now used the same definition of this variable throughout the manuscript (Lines 36, 338, 416, 562).

38) l. 325, 338, 344, 346 and later occurrences: overflow -> emission, release or similar.Check with native speaker.

Response:

We have replaced the inappropriate words in the manuscript (Lines 348, 362, 383, 485, 486).

39) l. 347: 'affecting the variation of the exchange fluxes' would be more precise in my view.

Response:

We have modified this sentence.

Line 385: "However, in general, WS, Chla, and [$C^{+}_{(s)tot}$] are the main factors affecting the variation of the exchange fluxes."

40) l. 349, 354: What is the temporal and spatial resolution of the model? This should be mentioned in section 2.3 where the model is described.

Response:

We have added a description of time resolution and grid resolution in section 2.3.

Line 172: "The time resolution is 1 hour, and the grid resolution is 27km×27km."

41) l. 358-360: Future tense ('will') does not sound correct.

Response:

We have corrected the error in tense.

Line 396: "The contribution of MBE to TMA and MMA effectively improved the regional simulation value."

42) l. 360-361: No significant change in autumn and winter seems to be the case for MMA and DMA only, according to Table 6.

Response:

We have now re-selected the simulation period in the process of revising the manuscript and have abandoned the expression of seasonal changes in the original manuscript.

43) l. 381 and other occurrences of ‚offshore': Shouldn't ‚offshore' be on the open ocean, at least farer away from the coast on the ocean? Why is land mass with 500m above sea level considered as ‚offshore'?

Response:

We have corrected the inappropriate words in the manuscript (Lines 281, 408, 425).

44) l. 393: Formulation sounds incorrect. Perhaps better 'was lowered / decreases by less than 10%'.

Response:

Thanks for the suggestion. In the process of revising the manuscript, we decided that Table 7 provided too little valuable information, and hence, the relevant expressions were deleted.

45) l. 396-398: Can that be shown with the model results, e.g. NO, OH concentration maps?

Response:

We have added the map and analysis of HONO and OH in the manuscript.

Lines 829-831:

“

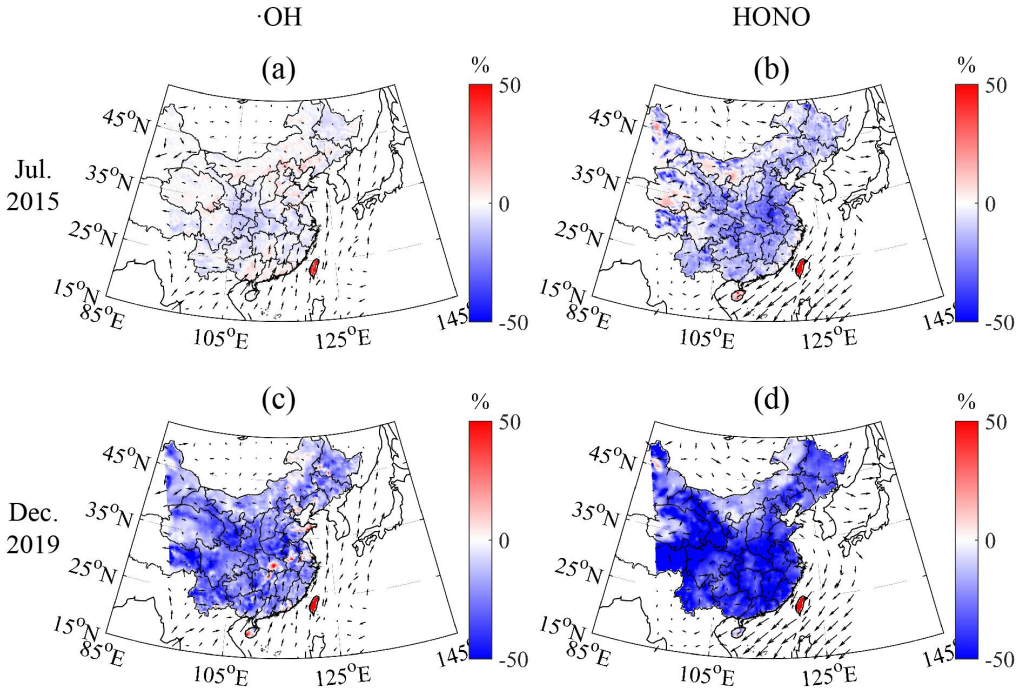


Figure 9: Spatial and temporal distribution of the change of ·OH and HONO mass concentration over land area. (a, c):·OH. (b, d): HONO.”

46) l. 400: What do the authors mean with ‚model mechanism‘?

Response:

We have now replaced the phrase with “model physical and chemical parameterization scheme.”(Lines 436-437)

47) l. 419-420: ‚they have relatively high fluxes’: What is the relative contribution of residential emissions in the mentioned areas? It seems that this is rather uniform and hence the relative change is more uniform (hence the relative change not as 'obvious' as you say).

Response:

We have modified the vague expression here.

Lines 456-459: “As can be seen from Fig. S1, residential emissions are uniformly distributed within a wide range. Although like agricultural emissions, they have larger emissions in areas such as the Sichuan Basin, North China Plain, Yangtze River Delta and Pearl River Delta, the relative difference is not obvious.”

48) l. 420-421: the difference ‘ -> If you refer to Fig. S2 and S3, the authors probably mean the relative difference ‘.

Response:

We have modified the inaccurate expression here.

Lines 456-459: “As can be seen from Fig. S1, residential emissions are uniformly distributed within a wide range. Although like agricultural emissions, they have larger emissions in areas such as the Sichuan Basin, North China Plain, Yangtze River Delta and Pearl River Delta, the relative difference is not obvious.”

49) l. 423: What is high value and low value area?

Response:

We have revised the vague expression here.

Line 461: “ there are significant differences between different regions”

50) l. 445-446: Shouldn't the change of pH due to Chla change directly affect the sea air exchange of DMA as well? What do you mean with 'the change of MBE of DMA is not considered'?

Response:

In the model emission files, the negative emission flux (i.e., sea is a sink) cannot be applied. Therefore, the influence of ocean sinks on DMA is not considered in this study.

51) l. 447: 'range of DMA' -> 'range of DMA change'

Response:

We have modified the text.

Lines 474-475: “ although DMA is less affected by MBE than MMA and TMA, dramatic changes in their concentrations still lead to changes of DMA concentrations.”

52) l. 449-450: therefore, DMA reacts with •OH faster than the other two amines in the upwind direction, and its concentration is less affected. ‘ Can the authors please explain in more detail what they mean here?

Response:

As mentioned above, only MBE of MMA and TMA are considered in this study, but the contribution of MBE to these two amines will affect the concentration of • OH in the atmosphere, and therefore indirectly affect the concentration of DMA. DMA is more easily oxidized by the • OH in the atmosphere as compared to the other amines. Therefore, the influence of changes in MMA and TMA concentrations is limited by their slow reaction rates, which cannot affect the concentration of •OH and DMA directly. Rather, it has a larger effect on downwind DMA concentrations over time. We have reorganized the content here in order to make the manuscript clearer.

Lines 500-504: “As mentioned above, only MBE of MMA and TMA are considered in this study, but the contribution of MBE to these two amines will affect the concentration of ·OH in the atmosphere, and thus indirectly affect the concentration of DMA. DMA is more easily oxidized by ·OH in the atmosphere as compared to the other amines. Therefore, the influence of changes in MMA and TMA concentrations is limited by their slow reaction rates, which cannot affect the concentration of ·OH and DMA directly.”

53) l. 466-467: going below 50 mm in October’ : What region does that refer to? Also in July many places in the domain have precipitation amounts < 50 mm.

Response:

We originally intended to express that the regions with precipitation <50mm in October are the most extensive compared with other months. We realized that this sentence is not rigorous and redundant, and have deleted it.

54) l. 475: DMA is only affected by terrestrial AE ‘: Didn't it have negative emission fluxes over sea water, i.e. is lost into the ocean surface (Fig. 4)?

Response:

In this study, marine sinks were not included in the parameterization scheme of the model, and hence, the analysis here indicates that DMA is mainly affected by terrestrial AE. We have revised the text accordingly.

Line 527: “In the study, there is only terrestrial AE of DMA....”

55) l. 476: variation range of its concentration -> variation range of its concentration change

Response:

We have modified the content.

Lines 528-529: “As a result, the variation range of its concentration over the sea surface is significantly higher than that of the other two amines”

56) l. 490: It's not the satellite data itself, but its application in an online emission scheme, that might reflect the emission situation.

Response:

We have modified the imprecise expression here.

Lines 546-547: “The emission scheme calculated from satellite data can better reflect the exchange direction of amines between ocean and atmosphere and reflect the emission characteristics of different amines.”

57) l. 494-495: Is that 500km away from the coast on land or 500km away over the ocean?

Response:

It is 500 km away from the coast. We have modified the vague expression here.

Lines 550-551: “The average increase rates of TMA in the two periods even reached 50% over 567 km and 378 km from the coastline.”

58) l. 495-496: These are the maximum increases for TMA due to MBE, aren't they? Why don’ t authors instead mention the average increases in the text as was done for MMA. I believe that more readers would be interested in averages than in the extremes?

Anyhow, it always must be clear what the given numbers are. This is missing throughout the manuscript in many places and makes it rather poor.

Response:

The TMA growth rate here is the average increase rate. We have revised the statement for clarity.

Lines 550-551: "The average increase rates of TMA in the two periods even reached 50% over 567 km and 378 km from the coastline."

59) l. 499: , WS and Chla were found to be the dominant factors affecting MBE fluxes': This is the case for all amines, right? Then please state it like this.

Response:

We have modified the expression here.

Line 562: "WS, Chla, and [$C^{+}_{(s)tot}$] were found to be the dominant factors affecting MBE fluxes for all of the above amines."

60) l. 507: , and the ocean also transforms from a source of amines to a sink': Was that observed in the simulations, since such result was not presented in section 3.3.2? And was it the case for all amines or only some? Since the Chla concentrations, according to Fig. 2, were mostly < 10 mg m-3, 50% increase should only make it a sink for DMA, and very slightly MMA perhaps.

Response:

This conclusion was mentioned in the results of the sensitivity experiments on the effect of Chla changes on amine exchange flux in Section 3.1 (Lines 346-347). We have now revised the statement.

Lines 566-567: "Chla indirectly influences the calculation results of exchange flux by affecting the calculation of pH."

61) Table 6: Some observation results are presented in μg m-3 and ng m-3. These should be transformed to pptv. Why are the Gao et al. 2022 observations of TMA much larger than for the other studies?

Response:

We have standardized the units, and marked those data whose units cannot be converted into pptv. In addition, we added a discussion on the data in Gao et al. (2020).

Lines 400-403: "In addition, it is worth noting that the TMA concentration observed by Gao et al. (2021) and Chen et al. (2021, 2022) is significantly higher than that in other literatures. This may be because TMA is mainly affected by MBE, and hence, concentrations of TMA observed by Gao et al. (2021) and Chen et al. (2021, 2022) at marine sites are much higher than those observed at terrestrial sites."

Lines 805-806: "

Table 6 Comparison of gaseous methylamines from simulations and measurement results in

different locations.

Location (Site Type)	Data Type	Date	MMA (ng m^{-3})	DMA (ng m^{-3})	TMA (ng m^{-3})	Ref.
Nanjing, China (Industrialized)	Measured	26 August - 8 September 2012	36.8*	77.5*	33.8*	Zheng et al. (2015)
Shanghai, China (Urban)	Measured	25 July - 25 August, 2015	19.7±7.4*	73.1±26.1*	2.6±1.4*	Yao et al. (2016)
Shanghai, China (Urban)	Measured	25 July - 25 August, 2015	19.7*	73.1*	2.6*	Mao et al. (2018)
	Simulated		6.3±8.3*	29.8±45.9*	2.4±3.3*	
Nanjing, China (Urban)	Measured	26-31 August 2012	5.5*	12.9*	4.6*	
	Simulated		8.0*	13.3*	1.4*	
Nanling Mountain, southern China (Background)	Measured	20 May - 9 June 2017	67.7±75.7*	211.1±156.0*	-	Liu et al. (2018)
		10 - 31 October, 2016	73.2±42.3*	86.3±65.8*	-	
Bohai Sea and Yellow Sea (Marine)	Measured	9 - 22 December 2019	-	6 ± 6	31±9	Gao et al. (2022)
	Measured	15 - 19 December, 2019		7±7	37±9	
The Yellow Sea (Marine)	Measured	7 – 16 January 2020	-	2.0±1.0	37±11	
East China Sea (Marine)	Measured	27 December 2019 – 7 January 2020	-	12±11	100±40	
The Yellow Sea and Bohai Sea (Marine)	Measured	December 2019 - January 2020	-	6.1±5.5	31.3±9.4	Chen et al. (2021)
the coastline of eastern China (Marine)	Measured	20 April - 16 May 2018	-	11±6.5	5.4±2.4	Chen et al. (2022)
Shanghai, China (Urban)	Simulated (Without MBE)	25 - 31 July 2015	1.3±0.5	2.8±1.6	0.5±0.2	This study.
The Yellow Sea and Bohai Sea (Marine)	Simulated (Without MBE)	9 - 22 December 2019	1.3±1.8	1.9±2.6	0.5±0.7	
Shanghai, China (Urban)	Simulated (With MBE)	25 - 31 July 2015	1.3±0.5	2.8±1.6	0.6±0.3	
The Yellow Sea and Bohai Sea (Marine)	Simulated (With MBE)	9 - 22 December 2019	1.4±1.9	2.1±2.7	6.1±3.5	

* Calculated from the data in references."

62) Fig. 2: The numbers in the plots: Black for SST, Chla, SSS and white color for WS would probably give better readability.

Response:

We have redrawn Fig.2.

Lines 809-811: “

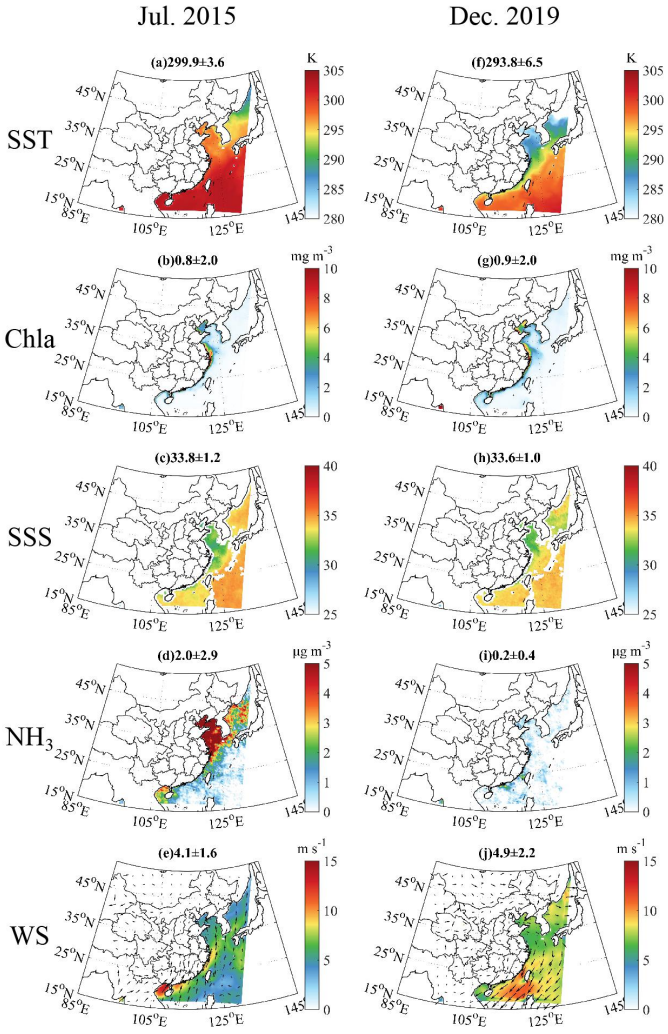


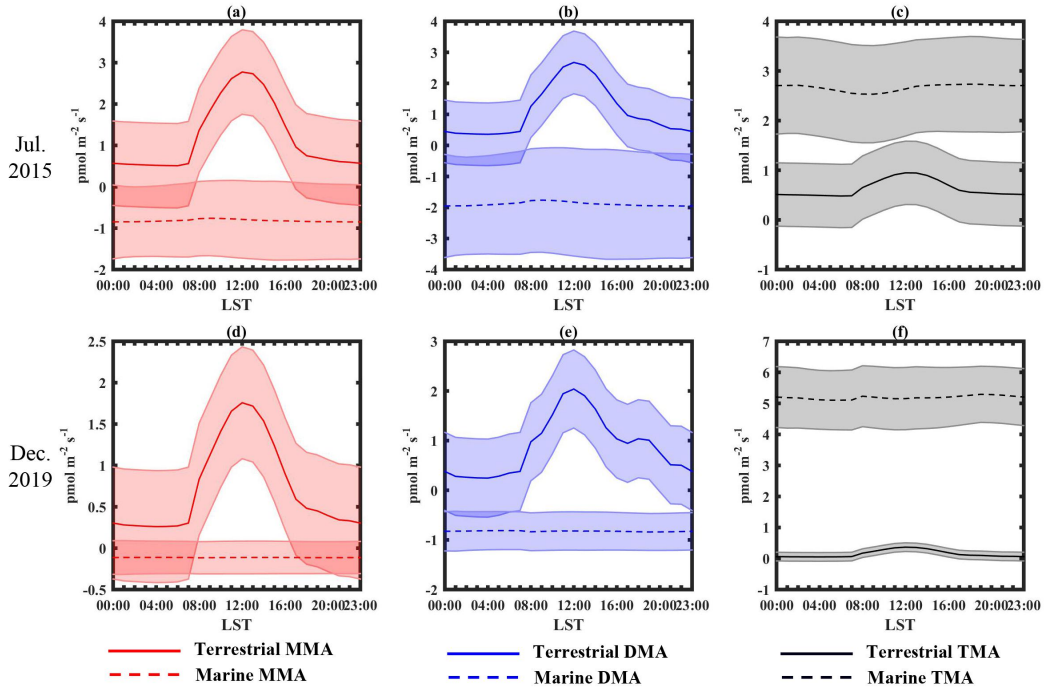
Figure 2: Temporal and spatial distribution of SST(a, f), Chla (b, g), SSS (c, h), NH_3 (d, i), and WS(e, j) used in simulation period. The numbers marked next to the serial number are the average.”

63) Fig. 4: Labels are wrong. In the figure, dashed is MBE and solid lines are AE.

Response:

We have corrected the labels in Fig. 4.

Lines 814-816:



“

Figure 4: Diurnal variation of amines emission fluxes average. (a, d) MMA, (b, e) DMA. (c, f) TMA. The solid lines represent the terrestrial AE fluxes, the dotted lines represent the MBE fluxes. The LST means the Beijing time(UTC+08:00).”

Comments to the author response to the quick initial review comments

1) In the response, changes in lines 362, 418, 464 are mentioned (with ，descriptions and data sources’), but I can’t find these changes.

Response：

We have re-marked the revised manuscript (Lines 390, 456, 519).

2) The comparison to measurements can of course be done only in a qualitative manner, butit should at least be discussed in the manuscript and not only presented in a table (Table 6).

Response:

We have added discussion on the data provided in Table 6.

Lines 396-406: “The contribution of MBE to TMA improved the regional simulation value. Table 6 shows the simulation results of this study and the observation and simulation results of other literature. Before adding MBE, the model significantly underestimated the concentration of amines in urban and marine sites. The addition of MBE resulted in a significant increase in the amines concentration simulated for marine sites (Fig. 7. 1116.7% for TMA and 9.0% for DMA.), effectively compensating for the model's underestimation. In addition, it is worth noting that the TMA concentration observed by Gao et al. (2021) and Chen et al. (2021, 2022) is significantly higher than that in other literatures. This may be because TMA is mainly affected by MBE, and hence, concentrations of TMA observed by Gao et al. (2021) and Chen et al. (2021, 2022) at marine sites are much higher than those observed at terrestrial sites. However, the simulated values at urban sites did not change significantly, indicating that the urban site is less affected by MBE in the simulation periods.

It is found that the AE inventory established based on MEIC in this study has a low resolution, and some strong emission sources are missing in the Yangtze River Delta where the urban site are located, which leads to the underestimation of the amines concentration at the urban site."

References

Bell. T.G., Johnson, M.T., Jickells, T.D., Liss, P.S.: Corrigendum to: Ammonia/ammonium dissociation coefficient in seawater: A significant numerical correction. Environ. Chem., 5, 258. https://doi.org/10.1071/en07032_co, 2008.

Chang, Y.H., Gao, Y.Q., Lu, Y.Q., Qiao, L.P., Kuang, Y.Q., Cheng, K., Wu, Y.H., Lou, S.R., Jing, S.A., Wang, H.L., Huang, C.: Discovery of a Potent Source of Gaseous Amines in Urban China, Environ. Sci. Technol. Lett., 8(9), 725-731, https://doi.org/10.1021/acs.estlett.1c00229, 2021.

Cree, C.H.L., Airs, R., Archer, S.D., Fitzsimons, M.F: Measurement of methylamines in seawater using solid phase microextraction and gas chromatography, Limnol. Oceanogr. Meth., 16, 411–420, https://doi.org/10.1002/lom3.10255, 2018.

Dall'Osto, M., Airs, R.L., Beale, R., Cree, C., Fitzsimons, M.F., Beddows, D., Harrison, R.M., Ceburnis, D., O'Dowd, C., Rinaldi, M., Paglione, M., Nenes, A., Decesari, S.,Simo, R.: Simultaneous Detection of Alkylamines in the Surface Ocean and Atmosphere of the Antarctic Sympagic Environment, A.C.S. Earth Space Chem., 3(5),854-+, http://dx.doi.org/10.1021/acsearthspacechem.9b00028, 2019.

Ge, X.L., Wexler, A. S., and Clegg, S.L.: Atmospheric amines e Part II. Thermodynamic properties and gas/particle partitioning, Atmos. Environ., 45, 561-577, https://doi.org/10.1016/j.atmosenv.2010.10.013, 2011b.

Gibb, S.W.: Trace Determination of Ammonia and Methylamines by Flow Injection Extraction: Ion Chromatography in Estuarine and Marine Environments, Ph.D., Tesis (Te University of East Anglia, Norwich), 1994.

Gibb, S.W., Mantoura, R.F.C., Liss, P.S.: Analysis of ammonia and methylamines in natural waters by fow injection gas difusion coupled to ion chromatography, Anal. Chim. Acta., 316, 291–304, https://doi.org/10.1016/0003-2670(95)00372-7, 1995.

Gibb, S.W., Mantoura, R.F.C., Liss, P.S., Barlow, R.G.: Distributions and biogeochemistries of methylamines and ammonium in the Arabian Sea, Deep Sea Res. Part II Top. Stud. Oceanogr., 46(3-4), 593-615, https://doi.org/10.1016/S0967-0645(98)00119-2, 1999b.

Gibb, S.W., Haton, A.D.: Te occurrence and distribution of trimethylamine-N-oxide in Antarctic coastal waters, Mar. Chem., 91, 65–75, https://doi.org/10.1016/j.marchem.2004.04.005, 2004.

Leng, C.B., Kish, J.D., Roberts, J.E., Dwebi, I., Chon, N., Liu, Y.: Temperature-Dependent Henry's Law Constants of Atmospheric Amines, J. Phys. Chem. A, 119(33), 8884-8891, https://doi.org/10.1021/acs.jpca.5b05174, 2015.

Li, M., Liu, H., Geng, G., Hong, C., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H., Man, H., Zhang, Q., He, K.: Anthropogenic emission inventories in China: a review, Natl. Sci. Rev., 4, 834-866, 2017.

Sander, R: Compilation of Henry's law constants (version 4.0) for water as solvent, Atmos. Chem. Phys., 15(8), 4399-4981, https://doi.org/10.5194/acp-15-4399-2015, 2015.

van Pinxteren, M., Fomba, K.W., van Pinxteren, D., Triesch, N., Hoffmann, E.H., Cree, C.H.L., Fitzsimons, M.F.,von Tumpling, W., Herrmann, H.: Aliphatic amines at the Cape Verde Atmospheric Observatory: Abundance, origins and sea-air fluxes, Atmos. Environ., 203, 183-195, https://doi.org/10.1016/j.atmosenv.2019.02.011, 2019.

Yang, X.H., Lee, C., Scranton, M.I.: Determination of Nanomolar Concentrations of Individual Dissolved Low Molecular Weight Amines and Organic Acids in Seawater, Anal. Chem., 65(5), 572-576, 1993.

Yang, X.H., Scranton, M.I., Lee, C.: Seasonal variations in concentration and microbial uptake of methylamines in estuarine waters, Mar. Ecol. Prog. Ser., 108(3), 303-312, https://doi.org/10.3354/meps108303, 1994.

Yu, G.R., Jia, Y.L., He, N.P., Zhu, J.X., Chen, Z., Wang, Q.F., Piao, S.L., Liu, X.J., He, H.L., Guo, X.B., Wen, Z., Li, P., Ding, G.A., Goulding, K.: Stabilization of atmospheric nitrogen deposition in China over the past decade, Nat. Geosci., 12(6), 424-+, https://doi.org/10.1038/s41561-019-0352-4, 2019.

Wu, C., Wen, Y., Hua, L., Jiang, J,m Xie, Y., Cao, Y., Chai, S., Hou, K., Li, H.: Rapid and highly sensitive measurement of trimethylamine in seawater using dynamic purge-release and dopant-assisted atmospheric pressure photoionization mass spectrometry, Anal. Chim. Acta, 1137, 56-63, https://doi.org/10.1016/j.aca.2020.08.060, 2020.