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 #1:

In the current study the authors provide a combined satellite and model study to estimate oceanic emissions of methylamine (MMA), dimethylamine (DMA) and trimethylamine (TMA) along the Chinese coastlines. Therefore, sea surface temperature (SST), chlorophylla (chl-a), sea surface salinity (SSS) and wind speed data were used. Recent investigations show that satellite data are a useful tool to simulate and understand emissions from the ocean. The study reveals that amine emissions from the ocean can have significant contribution to gas-phase TMA and MMA concentrations, but not to DMA concentrations.

Through sensitivity studies wind speed and chl-a concentrations were found to be important drivers of amine emissions. The modeled gas-phase concentrations of MMA, DMA and TMA are compared with measurements in that region and found good matches. Regarding the importance of amines for new particle formation and current limitations the paper addresses relevant scientific questions in the field of atmospheric chemistry.

Nevertheless, emissions from the oceans towards the atmosphere require good established concentration measurements within the sea water. These are not given for the Chinese coastline and thus the authors used an average value derived from different measurements. Here, high uncertainties can exist. Furthermore, the authors do not use established measured physical and chemical parameters for these amines. Besides, it seems that there is a bug in the calculation of the pKa value. Therefore, the simulations have to be reperformed.

Response:

According to the suggestions of reviewer, we have reperformed the simulations with corrected parameters and revised the whole manuscript accordingly. We mainly modified the calculation methods of the Henry's Law constant and pKa, and have added satellite-derived NH_3 column concentration to obtain a more correct and rigorous algorithm. Based on the above modification to the original algorithm, the new results show that the flux of amines from the atmosphere to the ocean slightly increases, while the flux from the ocean to the atmosphere slightly decreases; however, the main conclusions of the manuscript does not change.

In addition, we have obtained observational data from marine site for verification of the simulation results, thereby improving the simulation performance of amines concentrations at different types of sites (urban and marine sites) with the addition of MBE.

The paper needs major revision before publication.

Main Comments:

1）Parameters such as pKa and Henry's Law coefficient are important to calculate the amine flux into the atmosphere. The authors use Henry's Law coefficients obtained for NH3 as it was done in recent studies. This approach is feasible if such values are not given in literature. However, these values are determined. Why are the authors not using the Henry's Law coefficients for MMA, DMA and TMA as provided in Sander (2015) and Leng et al. (2015)? The pKa values used in table 4 from Gibb et al. (1999) are valid for 25°C not 20°C. Why are the temperature dependent pKa values given in the review of Ge et al. (2011b) not used? The simulations have to be reperformed by using the values for MMA, DMA, and TMA.

Response:

We thank the reviewer for the constructive comments. To clarify, we actually used pKa and Henry's Law coefficient for MMA, DMA, and TMA instead of NH_3 in our original manuscript but the temperature and ionic strength dependence of those parameters were adopted from NH_3. In the revised manuscript, the temperature dependence of pKa and Henry's Law coefficient have been updated using the values for amines. However, when calculating the ionic strength dependence of pKa we used the relation for NH_3 (Equation 9 in the revised manuscript) because such relation for amines is not available yet. The details of the revision are described below.

① Firstly, we have updated the calculation of the Henry's Law and associated constants. The new algorithm calculates the temperature-dependent Henry's Law coefficients based on the molar enthalpy of dissolution ($\Delta_{sol}H$, J mol $^{-1}$) of amines instead of NH_3.

Lines 196-200: "In this study, H of amines was also calculated based on Eq. (2) (Sander et al., 2015).

$$H(T) = H^{\ominus} \times \exp\left[\frac{-\Delta_{sol}H}{R}\left(\frac{1}{T} - \frac{1}{T^{\ominus}}\right)\right] \tag{2}$$

Where $H(T)$ is the value of H at the specified temperature T (K), $H^{\ominus}$ is H at standard temperature (298.15K), $\Delta_{sol}H$ (J mol $^{-1}$) is the molar enthalpy of dissolution, and R (8.314 J mol^{-1} K^{-1}) is the gas constant."

② We have also modified the reference temperature in the original manuscript from 20°C to 25°C.

Lines 238-239: "

$$pK_a = pK_{wa} + (0.1552 - 0.0003142T)I \tag{9}$$

where, pK_{wa} is the value of pK_a in the pure water (T=25 °C, I=0 mol dm^{-3}). ”

Lines 801-802:”

Table 4 Physicochemical parameters used in flux calculations.

Variable (Units)	Species			References
	MMA	DMA	TMA	
MW (g mol^{-1})	31.06	45.12	59.11	Gibb et al. (1999)
Henry’s law constant, H (T=25 °C, I=0 mol dm^{-3})	0.00055	0.00080	0.0040	Sander (2015); Leng et al. (2015)
Aqueous dissociation equilibria of atmospheric amines (first second acid dissociation constants), K_{wa} (T=25 °C, I=0 mol dm^{-3})	2.1878×10^{-11}	1.8621×10^{-11}	1.5849×10^{-10}	Ge et al. (2011b)
Δ_rH_0 (kJ mol^{-1})	53.737	49.450	36.017	
K_H (M atm^{-1}, T=25 °C)	89.3*	53.7*	8.9*	Sander (2015); Leng et al. (2015)
$\frac{d\ln H}{d(1/H)} = \frac{-\Delta_{sol}H}{R}$	4050*	5200*	5966*	

* Calculated from the data in references.”

③ In addition, we have also modified the calculation of pK_a. We replaced pK_{wa}, which was a constant in Eq. (9) in the original manuscript, with a variable varying with temperature, and at the same time added a related expression in the revised manuscript (also refers to comment 3 by Reviewer 1).

Lines 239-244: “Moreover, Bell et al. (2008) pointed out that K_{wa} in Eq. (9) should not be a constant, but should depend on temperature. The equation for calculating K_{wa} is as follows (Ge et al., 2011b):

$$\ln\left(K_{wa}(T)\right) = \ln\left(K_{wa}(T_r)\right) - \Delta_r H^0\left(\frac{1}{T} - \frac{1}{T_r}\right)/R \qquad (10)$$

Where $K_{wa}(T)$ is the value of the equilibrium constant at the specified temperature T (K), T_r is the reference temperature of 298.15 K, $\Delta_r H^o$ (kJ mol^{-1}) is the enthalpy change for the reaction at the reference temperature.”

2） The authors use observed values of MMA, DMA and TMA dissolved in sea water from other sea areas and state that “all sites are located in densely populated areas”. However, values from Hawaii or the Arabian Sea are used which are obviously not as densely populated areas as the Chinese coastline. Why do the authors use only these values, but neglect other measured values from Yang et al. (1994), Gibb et al. (1999), and van Pinxteren et al. (2019)?

Response:

We thank the reviewer for the constructive comments. We have added other measured values from recent publications to Table 3. In addition, we have also explained specific usage of the data in the table.

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 values 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.”

Lines 799-800: “

Table 3 A brief summary of previous research results on the distribution of amines.

Ocean	Location	Type	Period	[NH_3(g)] (pmol m^{-3})	[MMA(g)] (pmol m^{-3})	[DMA(g)] (pmol m^{-3})	[TMA(g)] (pmol m^{-3})	[$NH_{3(s)\ tot}^{+}$] (nM)	[$MMA_{(s)\ tot}^{+}$] (nM)	[$DMA_{(s)\ tot}^{+}$] (nM^3)	[$TMA_{(s)\ tot}^{+}$] (nM)	Ref.
Pacific	Hawaii	coastal	July–August 1985	-	11±5	93±51	30±19	-	52±20	1.5±20	12±3	Van Neste et al. (1987)
Atlantic	Narragansett, Rhode Island	coastal	July–August 1985	-	52±12	240±40	100±40	-	-	-	-	
Atlantic	Massachusettes	coastal	November 1984	-	-	-	-	-	200±58	8.9±4.4	41±27	
Atlantic	Massachusettes	coastal	November 1985	-	-	-	-	-	32±5	8.9±1.1	10±13	
Atlantic	Salt Pond	coastal	November 1985	-	-	-	-	-	55±20	6.7±2.2	10±4	
Atlantic	Flax Pond, New York	seawater	-	-					5-60	15-180	<3-80	Yang et al.,1993
Atlantic	Flax Pond, New York	seawater	-	-					5-40	25-180	10-50	Yang et al.,1994
Atlantic	Mediterranean	offshore	-	-				33±9.6	7.5±5.5	4.6±3.0	1.4±1.6	Gibb et al., 1994
Atlantic	Mediterranean	coastal	-	-				252±506	18±10.0	12±11.4	10±6.9	

Atlantic	Plymouth Sound, UK	coastal	-	-				230-594	4-23	13-22	4-17	Gibb et al., 1995a
Atlantic	Sutton Harbour	coastal	-	-				7700	91	<	15	
Atlantic	Mediterranean	offshore	-	-				22-60	<-9	<-9	<-7	
Atlantic	Mediterranean	coastal	-	-				26-660	4-38	3-15	4-22	
Atlantic	Arabian Sea	coastal	27 August–4 October 1994 (AS series)	(346-1726)946	37-177 (114)	16-65* (39)	<1od - 1.4(0.9)	80-150 (108)	<3od-658 (16.2)	<lod-13.9 (4.54)	<lod-0.44 (0.044)	Gibb et al., 1999a
Atlantic			16 November -19 December 1994 (AS series)	(2454-5628)3780	50-241 (143)	50-870* (196)	<1od-1.4 (8.1)	81-253 (156)	10.1-49.8 (25.6)	<lod-9.95 (4.85)	<lod-0.8 (0.14)	
Atlantic	Arabian Sea	offshore	27 August–4 October 1994 (A series)					70-150 (115)	<lod-13.9 (5.24)	<lod-11.1 (3.85)	<lod (n/a)	
Atlantic			16 November -19 December 1994 (A seires)					137-230 (230)	<lod-16.5 (8.8)	<lod-5.7 (2.04)	<lod-0.3 (0.036)	
Atlantic	Arabian Sea	coastal	August–October 1994 (AS series)					139±135	12±20	3.0±4.1	0.10±0.37	Gibb et al., 1999b
Atlantic			November -December 1994 (AS series)					206±271	22±13	4.2±2.8	0.45±0.81	
Atlantic	Arabian Sea	offshore	August–October 1994 (A series)					91±91	6±7	2.9±2.8	0.05±0.21	
Atlantic			November -December 1994 (A seires)					112±76	12±7	2.9±1.6	0.13±0.24	

Atlantic	Arabian Sea	coastal	August–October 1994 (GOM series)						113±102	11±9	2.8±3.1	0.19±0.42	
Atlantic	Ryder Bay	seawater	January – February 1999							12.0±9.1	3.8 ± 3.9	1.6 ± 1.8	Gibb and Hatton, 2004
Atlantic	Western English Channel	seawater	January – February 1999							3	6	20	Cree et al., 2018
Atlantic	the island of Sao Vicente	seawater	November 2011 November 2013							5-33 11-23	2-15 7-197	- -	van Pinxteren et al., 2019
Atlantic	the regions of Antarctic Peninsula, South Orkney, and South Georgia Islands	seawater	January-February, 2015									4.2	Dall'Osto et al., 2019
Pacific	the coastal zones of Dalian	seawater										8.3 3.7 5.1 6.3 3.1 3.2	Wu et al., 20s20

”

3) Formula 10 In the publication of Khoo et al. (1977) the formula is different. It is $pKa = pKwa + (0.1552 – 0.0003142T)*I$ instead of the applied $pKa = pKwa + (0.1552 – .003142T)*I$ Furthermore, there was a correction of this prediction method by Bell et al. (2007; 2008). This has to be checked and the simulations have to be reperformed.

Response:

Thanks to the reviewer for the careful scrutiny. We have corrected the mistake in this equation. In addition, according to the modified method in Bell et al. (2008), the pK_{wa} calculation in this equation is supplemented. According to Bell et al. (2008)'s verification of Khoo et al. (1977)'s prediction method, pK_{wa} is not a constant at a certain reference temperature, but should vary with temperature. We adopt the method for calculation of temperature dependence of pK_{wa} in Ge et al. (2011b). Corresponding instructions have been added to the manuscript.

Lines 236-244: "

Further, K_a is also dependent on the temperature and ionic strength. For the $NH_{3(s)}$–$NH_4^+{}_{(s)}$ system, 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.

$$pK_a = pK_{wa} + (0.1552 - 0.0003142T)I \qquad (9)$$

Where pK_{wa} is the value of pK_a in the pure water (T=25 °C, I=0 mol dm^{-3}). Moreover, Bell et al. (2008) pointed out that K_{wa} in Eq. (9) should not be a constant, but should depend on temperature. The equation for calculating K_{wa} is as follows (Ge et al., 2011b):

$$\ln(K_{wa}(T)) = \ln(K_{wa}(T_r)) - \Delta_r H^0 \left(\frac{1}{T} - \frac{1}{T_r}\right) / R \qquad (10)$$

Where $K_{wa}(T)$ is the value of the equilibrium constant at the specified temperature T (K), T_r is the reference temperature of 298.15 K, $\Delta_r H^o$ (kJ mol^{-1}) is the enthalpy change for the reaction at the reference temperature."

In addition, satellite-derived NH_3 column concentration was added in this study. Combined with the algorithm modification of comments (1) and (2), and comparing with the calculation results in the original manuscript, the final MBE fluxes showed that the flux of amines exchange from the ocean to the atmosphere slightly decreased, while the flux from the atmosphere to the ocean slightly increased. This observation, however, does not significantly change the final conclusion.

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.

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

$[NH_3]_G$ 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,

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

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

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

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

Lines 803-804: “

Table 5 MBE fluxes of the three types of amines over ocean in July 2015 and December 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)

”

Lines 812-813: “

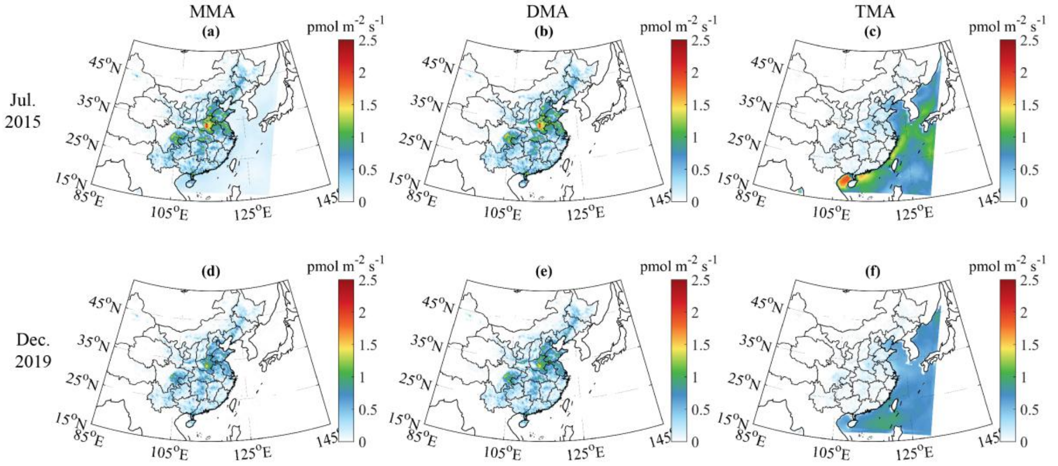


Figure 3: Spatial and temporal distribution of methylamines emissions: (a, d) MMA; (b, e) DMA; (c, f) TMA.”

4) The authors state that chl-a influences the emission of amines into the atmosphere, but concentrate only on the pH effect. In environments rich of biological activity such as the sea-surface microlayer DMA concentrations can be up to one order of magnitude larger than in the bulk (van Pinxteren et al, 2019). A sensitivity study dealing the

possible effect of higher chl-a on dissolved amine concentrations is missing and has to be done.

Response:

Thanks for the constructive comments. The biochemical activity in the sea-surface microlayer does have a significant effect on the dissolution of amines in seawater. However, many of the biochemical processes involved in these processes are complex, and hence, there is still no effective algorithm for quantitative analysis of these processes. Therefore, only the intermediate variable pH in the empirical equation (Eq. (7)) involving Chla in this study was analyzed to assess the effect of Chla on amine dissolution. We have also added a sensitivity study about the synergistic effects of Chla, SST, and SSS on pH, pKa, and the concentration of unionized amine molecules.

Lines 345-372: "

(b) Chla

With the increase of Chla, the direction of amines exchanges between the ocean and atmosphere showed a trend of transferring from the atmosphere to the ocean (Fig. 5 (b, i)). According to Eq. (7), the increase of Chla will lead to the decrease of pH (Fig.6(a, e)), and hence, it is not conducive to the emission of amines. Chla is used to indicate primary production in the water body, and high Chla indicates a significant increase in phytoplankton. Water eutrophication is caused by the massive growth of phytoplankton due to the continuous importation of anthropogenic nutrients into the sea by rivers. Thus, the increased organic matter is transported to the subsurface water by settling and being decomposed by microorganisms. This process consumes oxygen in the water and forms a hypoxic environment. With the mixing of the water, the pH of the water changes. Zhao et al. (2020), based on the observation data of summer voyage from the Pearl River Estuary to the northern continental shelf of the South China Sea, found that the water on the west side of the Pearl River Estuary with obvious mixing of fresh water and seawater is characterized by low dissolved inorganic carbon (DIC) and high pH, while for the area with 20-30 meters water depth outside the mixing area, it is characterized by high DIC and low pH. The main source of amines comes from the degradation of organic matter in sediments (Carpenter et al., 2012), and therefore, an increase in Chla might mean more acidification in the ocean, making amines more soluble in seawater.

(c) SSS

It can be seen from Fig. 5 (c, j) that the increase of SSS will increase the tendency of the amines to be emitted from the water surface. It can be inferred from Eq. (8), (9), (10), and (11) that the increase of SSS will further inhibit the ionization of amines in seawater (Fig. 6((b, f)), which makes it more prone to emission from the water surface, resulting in an increase of the exchange fluxes.

(d) SST

An increase in SST will lead to a decrease of the fluxes of amines emitted from the ocean to the atmosphere (Fig. 5 (d, k)). As can be seen from Eq. (7), the increase of SST will lead to the decrease of pH (Fig.6(a, e)) and the reduction of amine *pKa* (Fig.6(b, f)), which makes the amines more likely to dissolve in water (Fig.6(c, g)). Due to the high stability of the marine environment, the variation range of SST itself is small; therefore, SST has the least influence on the exchange flux among the four elements. It should be noted that El Niño conditions occurred by late May in 2015, which increased the global average temperature and affected the weather patterns in the study area (Kennedy et al., 2016). The annual average SST of our study areas was 0.5-1.0°C higher than the average value recorded during the period from 1961 to 1990 (Kennedy et al., 2016). Figure 5(d ,k) shows that the exchange fluxes of amines are negatively correlated with SST, but the sensitivity is low."

Lines 817-820: "

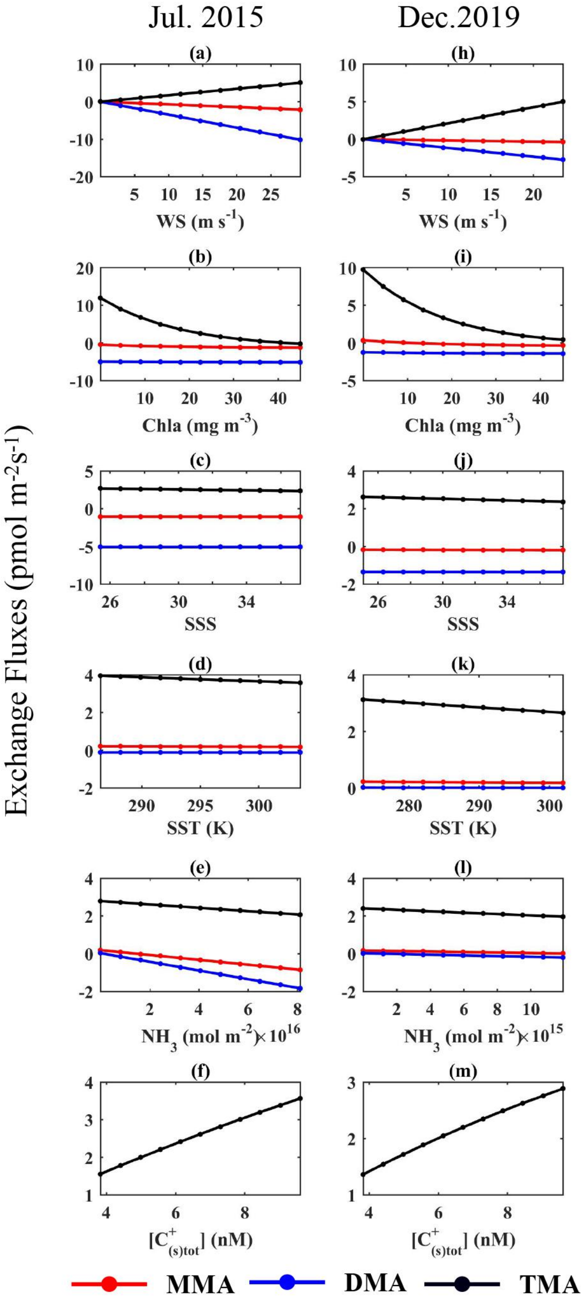


Figure 5: Influence of WS (a, h), Chla (b, i), SSS (c, j), SST (d, k), NH$_3$ (e, l), and [C$^+_{(s)tot}$] (f, m) on the average of MBE. In order to ensure the simplicity of the figure, only the changes of TMA are plotted here, and the changes of the other two amines are consistent with TMA."

Lines 821-823: “

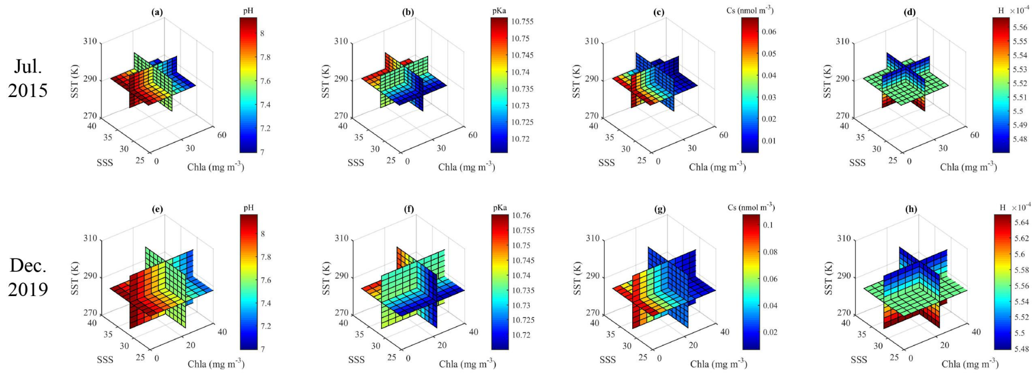


Figure 6: Influence of Chla, SSS and SST on pH (a, e), pKa (b, f. In the case of MMA), Cs (c, g. In the case of MMA), and Henry’s Law coefficient (d, h. In the case of MMA).”

5) For anthropogenic emissions the authors use the Amines-to-Ammonia mass emission ratio derived for the Yangtze River Delta region. This ratio will result into lower TMA emissions compared to MMA and DMA. However, in the review of Ge et al. (2011a) animal husbandry TMA emissions dominate MMA and DMA that is not reflected in the current study by the lower agricultural emission ratio. Thus, the high ocean contribution might be coincidence because of the underestimation. The ratio for agricultural emission from Mao et al. (2018) is further 0.00043 instead of the applied 0.0004. Besides, recently a new source of C2 and C3 amines was detected in China (Chang et al., 2021) that might be a potent TMA source. This is not represented, yet.

Response:

We strongly agree with the reviewer's view that animal husbandry and other emissions are important sources of amines, but they have not been included in our emission inventories. Chang et al. (2021) have tried to establish a grid inventory of DMA and TMA for septic tanks in the Yangtze River Delta region, but there are still great uncertainties in the establishment of agricultural emission inventory for a larger domain. Due to the complex factors affecting agricultural production (e.g., fertilizer application methods, animal feeding conditions and manure management) and the different environmental conditions (e.g., surface temperature, soil acidity, soil moisture content and wind speed) that determine ammonia volatilization (Li et al., 2017), it is difficult to accurately estimate ammonia and amine emissions nationwide.

Therefore, this study applied the ratio for agricultural emission from Mao et al. (2018). In addition, the data we used when establishing the anthropogenic emission inventories of amines was consistent with Mao et al. (2018). Table 1 contained a numerical error; however, it did not affect the simulation results in the manuscript, and we have corrected the errors in the revised manuscript. We have also added a discussion on anthropogenic source uncertainties in the manuscript to make it clearer.

Lines 138-145: “For example, animal husbandry and other agriculture emissions have been important sources of amines, but they have not been included in our emission inventories.

Due to the complex factors affecting agricultural production (e.g., fertilizer application methods, animal feeding conditions and manure management) and the different environmental conditions (e.g., surface temperature, soil acidity, soil moisture content and wind speed) that determine ammonia volatilization (Li et al., 2017), it is difficult to accurately estimate ammonia and amines emissions nationwide. Chang et al. (2021) have tried to establish a grid inventory of DMA and TMA for septic tanks in the Yangtze River Delta region, but there are still great uncertainties in the establishment of agricultural emission inventory for a larger region."

6) Why do the authors not treat uptake on aerosols that was determined to be important for the lifetime of amines by Yu and Luo (2014)?

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."

7) The authors do refer to a mass ratio of amines with ammonia from Zheng et al. (2015) for the chemical boundary conditions of the model, but there a ratio between amines as well as ammonia with NOx is given. From the values of Zheng et al. (2015) it is hard to recalculate the ratio presented in this study.

Response:

We thank the reviewer for the careful scrutiny. The mass ratio of amines with ammonia adopted in this study refers to the ratio from the Fig. 7 of Zheng et al. (2015), which was obtained from the observed concentration values of atmospheric amine and ammonia during the whole study period. However, the ratio between amines as well as ammonia with NOx provided by Zheng et al. (2015) is the ratio in the smoke plume of the factory. Moreover, it can be seen from Fig. 7 of Zheng et al. (2015) that the ratio of amines to NOx in the plume is different from that in the actual atmosphere.

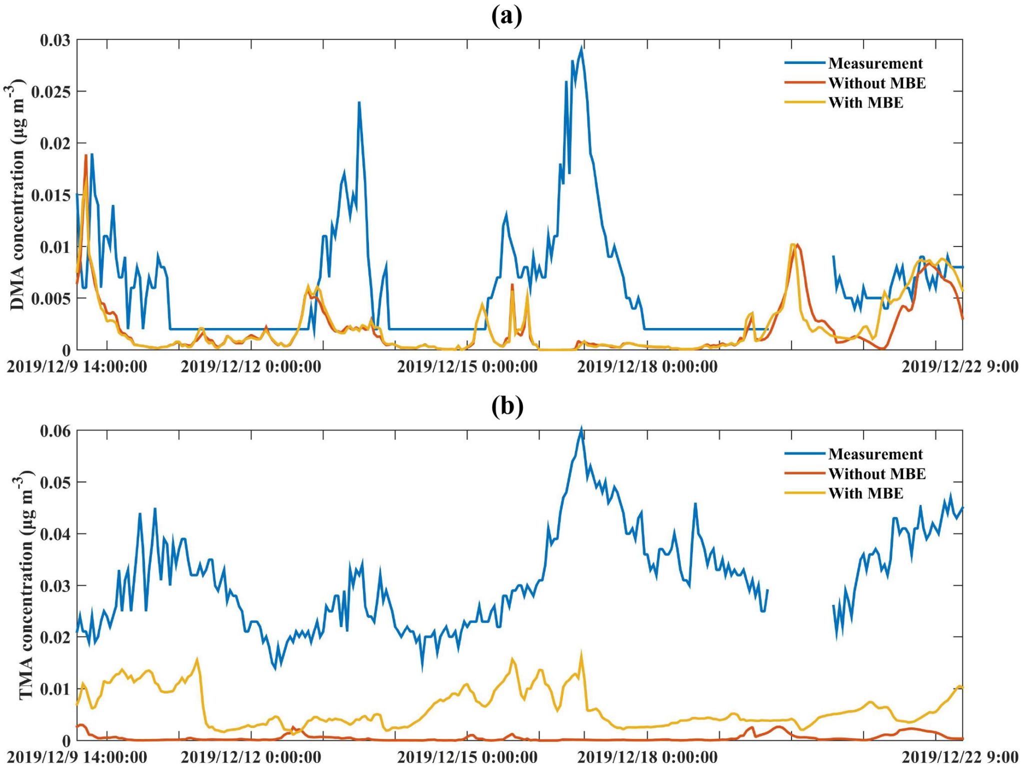
8) There a two recent measurements campaigns of marine MMA, DMA and TMA in the gas phase at the study area. However, it seems the results are not well discussed. For example, TMA measured is around one order of magnitude higher in Gao et al. (2022), but a discussion is missing.

Response:

As suggested, we have added a relevant discussion 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.”

9) Furthermore, in table 6 the values are sometimes in ng or µg m-3 instead of pptv making a comparison difficult. I suggest that the authors include their modeled average values together with pptv also in ng m-3 for better comparison. Overall, from the most recent studies it seems that during winter the model overpredict DMA, but underpredict TMA.

Response:

Thanks for the suggestion. For the convenience of comparison, we have converted all units of pptv in literature to ng m^{-3} and our simulated results also shown with the unit of ng m^{-3} (Table 6).

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	19.7*	73.1*	2.6*	Mao et al.

		August, 2015				(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.”

Minor Comments

1) Line 61 Does this refer towards NH3? Should be better specified.

Response:

We have improved the 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).”

2) Line 93 Delete “And”

Response:

Done.

3) Line 104 “Multiple”

Response:

Done.

Lines 114-118: “ Therefore, we attempt to establish an emission inventory of amines including MMA, DMA, and TMA from MBE based on their mechanisms of production. 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.”

4) Lines 108-109 The authors focus only on a very small part of the North Pacific. The sentence should be rephrased.

Response:

Thanks for the reviewer's suggestion. We have modified the imprecise expression here.

Lines 121-122: “The research area of this study is shown in Fig. 1, which mainly explores the contribution of MBE to atmospheric amines concentration in southeast China.”

5) Line 142 Please provide a reference for the reaction rate coefficients.

Response:

Here we no longer use the reaction rate coefficients of Ge et al. (2011a) quoted in the original manuscript, but instead use those of Carl and Crowley (1998). Because the reaction rate coefficients quoted in the original manuscript are derived from Atkinson et al. (1977, 1978) quoted in Ge et al. (2011a), compared with the results of Carl and Crowley (1998), it is older.

Lines 162-163: “the hydroxyl radical reaction rate constants (K_{OH}, cm^3 mol^{-1} s^{-1}) were 1.79×10^{-11}, 6.49×10^{-11}, and 3.58×10^{-11} for MMA, DMA, and TMA, respectively (Carl and Crowley, 1998).”

6) Line 145 How long was the spin-up time for the model?

Response:

The spin-up time for the model is 64 hours. We have also added relevant content to the original manuscript to make the description of the setup of the model clearer.

Line 177: “The spin-up time for the model is 64 hours.”

7) Line 255 From the figure it seems that it is April > October > July > January.

Response:

We 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, therefore, re-selecting 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.

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: “

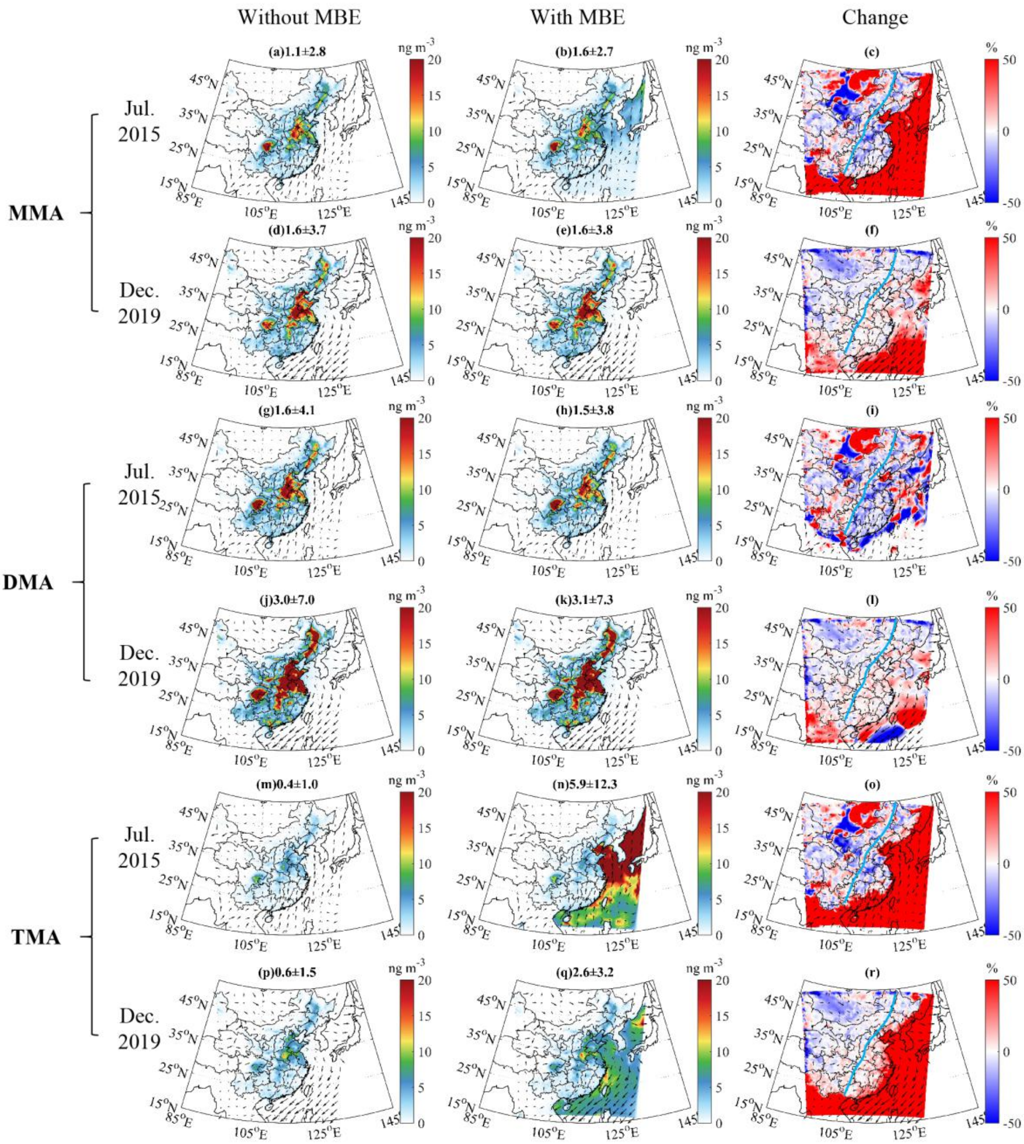


Figure 8: Spatial and temporal distribution of amines simulated mass concentration. (a-f): MMA. (g-l): DMA. (m-r): TMA. The solid blue line represents the boundary between the second and third steps of the Chinese terrain."

8) Line 257 "to the at the ocean" rephrase

Response:

We have corrected the grammatical errors here.

Line 281: "As can be seen from Fig.2(b, g), the coastal area exhibits high values of Chla concentration."

Lines 809-811: "

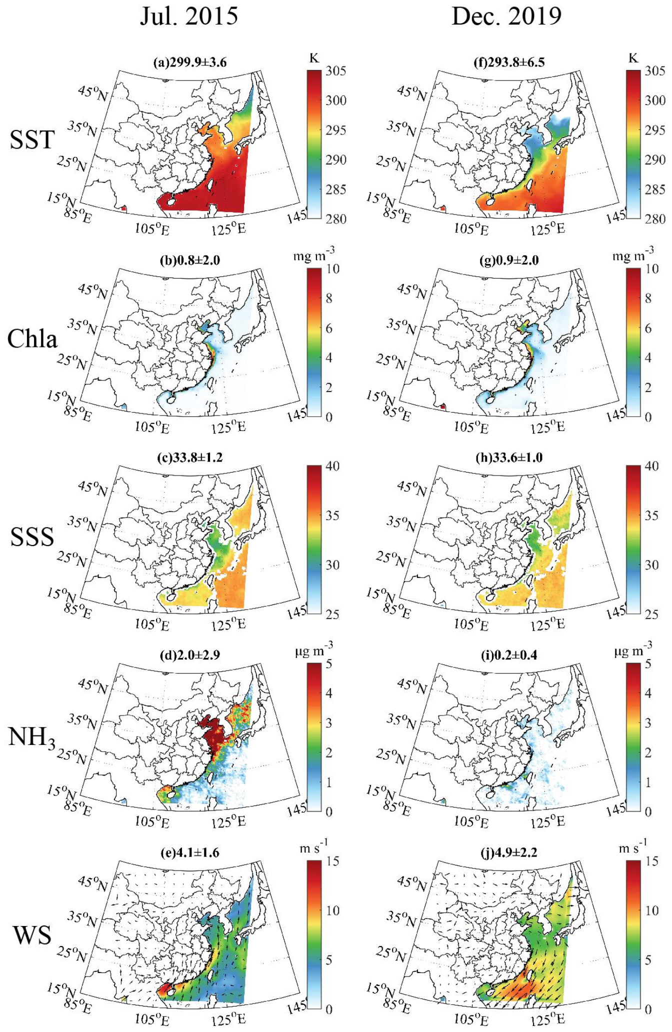


Figure 2: Temporal and spatial distribution of SST(a, f), Chla (b, g), SSS (c, h), NH_3 (d, i), and WS(e, j) used in simulation period. The numbers marked next to the serial number are the average."

9) Line 302 Is this the average of the full area? Has to be mentioned.

Response:

Modified as suggested.

Lines 322-323: "The area-averaged marine emission fluxes of the three types of amines are shown in Table 5."

10) Line 303 Is the last part really needed?

Response:

We deleted the redundant part.

11) Line 311 The referring to MBE is missing for the TMA comparison.

Response:

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 calculated MBE fluxes was deleted.

12) Line 334 Provide a reference for the degradation in sediments.

Response:

We have added a reference here (Line 355).

13) Line 342 With decreasing SST the emission increases.

Response:

We have corrected the error of expression here.

Line 372: "Figure 5(d ,k) shows that the exchange fluxes of amines are negatively correlated with SST, but the sensitivity is low."

14) Section 3.2 For the reader it would be better when the discussion is structured into (i) MMA, (ii) DMA and (iii) TMA as it is for the figures. Separate figures showing the percentage changes over the ocean will help to better understand and follow the discussion.

Response:

Thanks for your constructive comments. We have restructured the discussion in Section 3.1 and 3.2.

Lines 337-384: "

(a) WS

As shown in Eq. (3), WS is directly used in the calculation of K_g, which is directly proportional to K_g, and has the most direct influence on the calculation of exchange fluxes. Higher WS will accelerate the material exchange between the ocean and the atmosphere, which is directly reflected in the change of the exchange fluxes, and the two show a significant linear relationship (Fig. 5(a, h)). For MMA and TMA, the ocean is the source, and the increase of WS will lead to a linear increase in the emission fluxes. For DMA, the ocean is a sink, and the increase in WS also accelerates the transportation of atmospheric DMA into the ocean. The other three variables have an indirect effect on the exchange fluxes mainly by affecting the calculation of intermediate variables.

(b) Chla

With the increase of Chla, the direction of amines exchanges between the ocean and atmosphere showed a trend of transferring from the atmosphere to the ocean (Fig. 5 (b, i)). According to Eq. (7), the increase of Chla will lead to the decrease of pH (Fig.6(a, e)), and hence, it is not conducive to the emission of amines. Chla is used to indicate primary production in the water body, and high Chla indicates a significant increase in phytoplankton. Water eutrophication is caused by the massive growth of phytoplankton due to the continuous importation of anthropogenic nutrients into the sea by rivers. Thus, the increased organic matter is transported to the subsurface water by settling and being decomposed by microorganisms. This process consumes oxygen in the water and forms a hypoxic environment. With the mixing of the water, the pH of the water changes. Zhao et al. (2020), based on the observation data of summer voyage from the Pearl River Estuary to the northern continental shelf of the South China Sea, found that the water on the west side of the Pearl River Estuary with obvious mixing of fresh water and seawater is characterized by low dissolved inorganic carbon (DIC) and high pH, while for the area with 20-30 meters water depth outside the mixing area, it is characterized by high DIC and low pH. The main source of amines comes from the degradation of organic matter in sediments (Carpenter et al., 2012), and therefore, an increase in Chla might mean more acidification in the ocean, making amines more soluble in seawater.

(c) SSS

It can be seen from Fig. 5 (c, j) that the increase of SSS will increase the tendency of the amines to be emitted from the water surface. It can be inferred from Eq. (8), (9), (10), and (11) that the increase of SSS will further inhibit the ionization of amines in seawater (Fig. 6((b, f)), which makes it more prone to emission from the water surface, resulting in an increase of the exchange fluxes.

(d) SST

An increase in SST will lead to a decrease of the fluxes of amines emitted from the ocean to the atmosphere (Fig. 5 (d, k)). As can be seen from Eq. (7), the increase of SST will lead to the decrease of pH (Fig.6(a, e)) and the reduction of amine *pKa* (Fig.6(b, f)), which makes the amines more likely to dissolve in water (Fig.6(c, g)). Due to the high stability of the marine environment, the variation range of SST itself is small; therefore, SST has the least influence on the exchange flux among the four elements. It should be noted that El Niño conditions occurred by late May in 2015, which increased the global average temperature and affected the weather patterns in the study area (Kennedy et al., 2016). The annual average SST of our study areas was 0.5-1.0°C higher than the average value recorded during the period from 1961 to 1990 (Kennedy et al., 2016). Figure 5(d ,k) shows that the exchange fluxes of amines are negatively correlated with SST, but the sensitivity is low.

(e) NH_3 and $[C^{+}_{(s)tot}]$

Figure 5 (e, l, f, m) shows that shows the relationship between the exchange fluxes and ammonia concentration and $[C^{+}_{(s)tot}]$. It can be seen that the increase of the former will promote gas from the atmosphere into the ocean, while the latter will promote gas from the ocean into the atmosphere. This situation is related to Eq.(1). According to the algorithm of this study, increasing ammonia concentration means increasing the concentration of amines in the atmosphere ($[C_{(g)}]$). When the concentration of amines in the atmosphere is large enough, the atmosphere acts as the source and the ocean acts as the sink. 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, Henry's law coefficient (*H*), as an important parameter affecting water-gas exchange, is also affected by marine environmental parameters. It can be seen from Fig. 6(d, h) that with the increase of SST and SSS, H will decrease, which means that amines are more inclined to emission from the sea water, which may change the direction of sea-air exchange of amines."

Lines 409-414: “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).”

15) Line 364 Table 7 instead of table 6

Response:

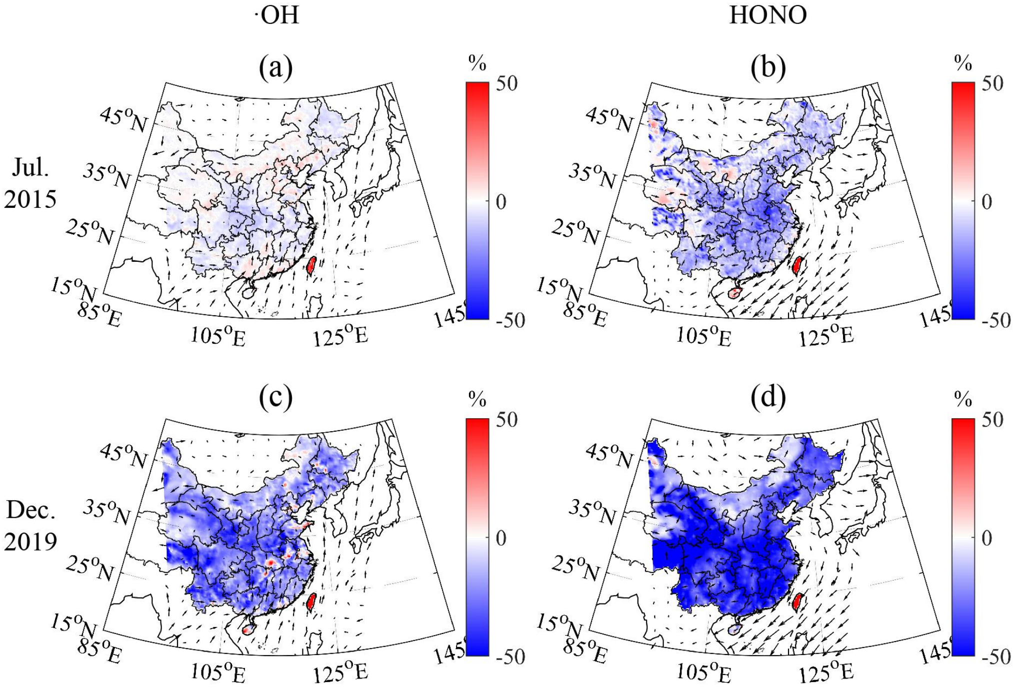
In the process of revising the manuscript, we decided that Table 7 provided too little valuable information, and hence, the relevant expressions were deleted.

16) Line 395 How is the difference of HONO + OH between the simulation with and without MBE?

Response:

We have supplemented the changes of HONO + OH to support the discussion 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.”

17) Line 405 Add that the low agricultural emissions of DMA are related to the contribution.

Response:

Thanks for the suggestion. We have added the corresponding content in the manuscript.

Lines 441-444: “The difference in AE composition indicates that different emission sources have different effects on different species of amines. As shown in Table S2, agricultural emission is the major source of the three types of amines. However, the contribution of agricultural emissions to DMA are relatively low compared with those of MMA and TMA (accounting for more than 80%). ”

18) Line 419 In figure S2 and S3 the distribution of the residential emissions is not shown.

Response:

We have corrected the misstatement here.

Lines 456-457: “As can be seen from Fig. S1, residential emissions are uniformly distributed in a wide range.”

19) Section 3.3.2 A discussion of DMS when the WS is reduced is missing. Why are there so strong changes in July?

Response:

We have added the missing discussion to make the article clearer.

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

The change in July is more intense because the prevailing winds in July blow from the sea to the land, bringing more marine influences (known as MBE). Therefore, when the MBE changes during the month, the impact is more dramatic. We have also added relevant content to the article to make the original discussion clearer.

Lines 476-477: “bringing more Marine influences (known as MBE). Therefore, when the MBE changes during the month, the impact is more dramatic.”

20) Section 3.3.3 When the Henry’s Law coefficient is changed also the emission flux is changed. This discussion is missing.

Response:

Thanks for the reviewer's suggestion. Henry's Law coefficient is actually one of the intermediate variables caused by the marine environmental parameters (SST, SSS, etc). We have added discussion on the impact of marine environmental parameters on Henry's Law coefficient, and how Henry's Law coefficient affects emission fluxes.

Lines 381-384: “In addition, Henry's law coefficient (H), as an important parameter affecting water-gas exchange, is also affected by marine environmental parameters. It can be seen from Fig. 6(d, h) that with the increase of SST and SSS, H will decrease, which means that amines are more inclined to emission from the sea water, which may change the direction of sea-air exchange of amines."

Lines 821-823: "

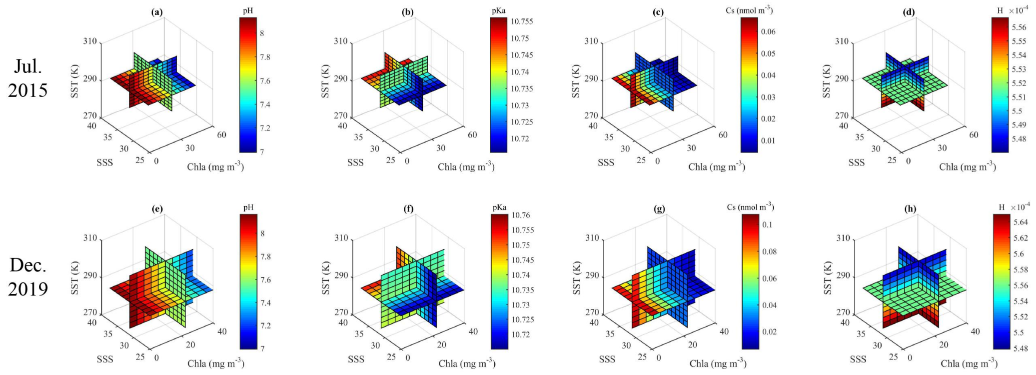


Figure 6: Influence of Chla, SSS and SST on pH (a, e), pKa (b, f. In the case of MMA), Cs (c, g. In the case of MMA), and Henry's Law coefficient (d, h. In the case of MMA)."

21) Line 491 Because of the missing sea water data and the missing uptake on aerosol particles, it could be questionable if the last part of the sentence ("which is more consistent with the reality") is really true. It might be true on global scale, but not necessarily on regional scale.

Response:

Thanks for the reviewer's suggestion, we have deleted this overly general conclusion.

22) Table 6 Please add the year of the measurement. Provide also average values.

Response:

We have added the year of the measurement and average values in the table. In addition, we deleted the amines concentration data of Yao et al. (2018), because the data corresponded to days with new particle formation events, which was special.

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."

23) Figure 4 The figure caption describes MBE and AE contribution different as mentioned in the figure legend.

Response:

We have corrected the error in the figure caption.

Lines 814-816: "

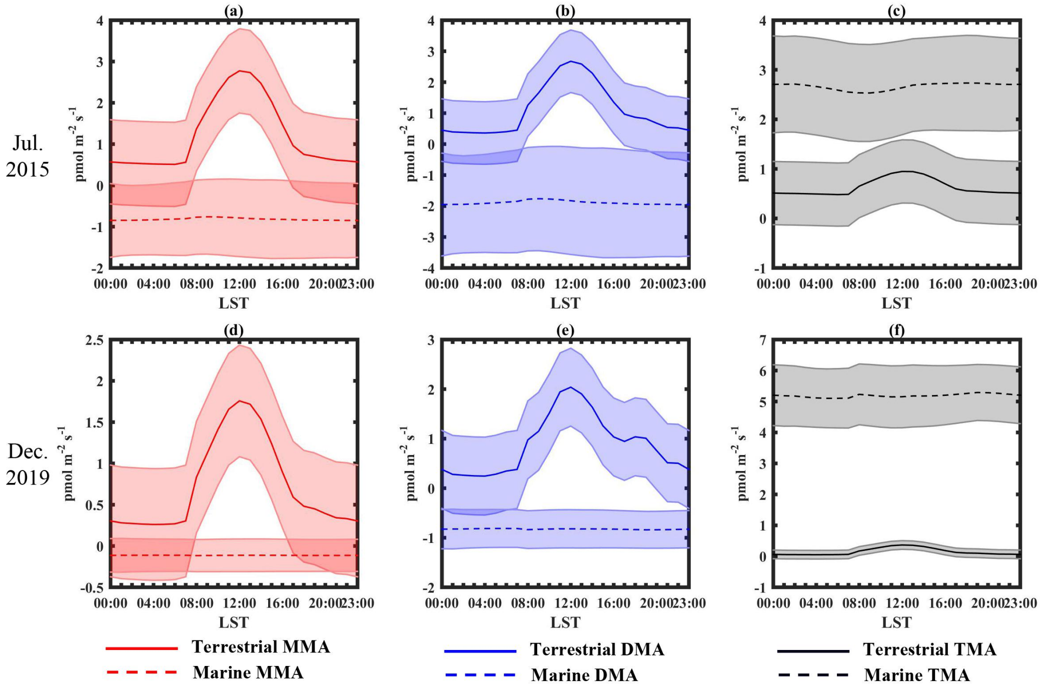


Figure 4: Diurnal variation of amines emission fluxes average. (a, d) MMA, (b, e) DMA. (c, f) TMA. The solid lines represent the terrestrial AE fluxes, the dotted lines represent the MBE fluxes. The LST means the Beijing time(UTC+08:00).”

24) Figure S2-S6 Give the name of month instead of a number and symbol.

Response:

We have modified accordingly.

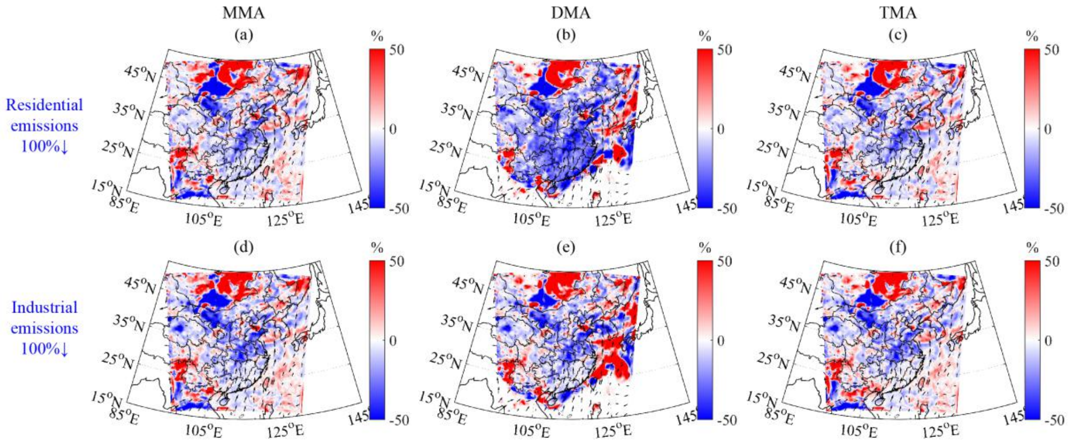


Figure S2: Changes in amines simulated concentration after reduction of residential emissions and industrial emissions (In the case of July 2015).

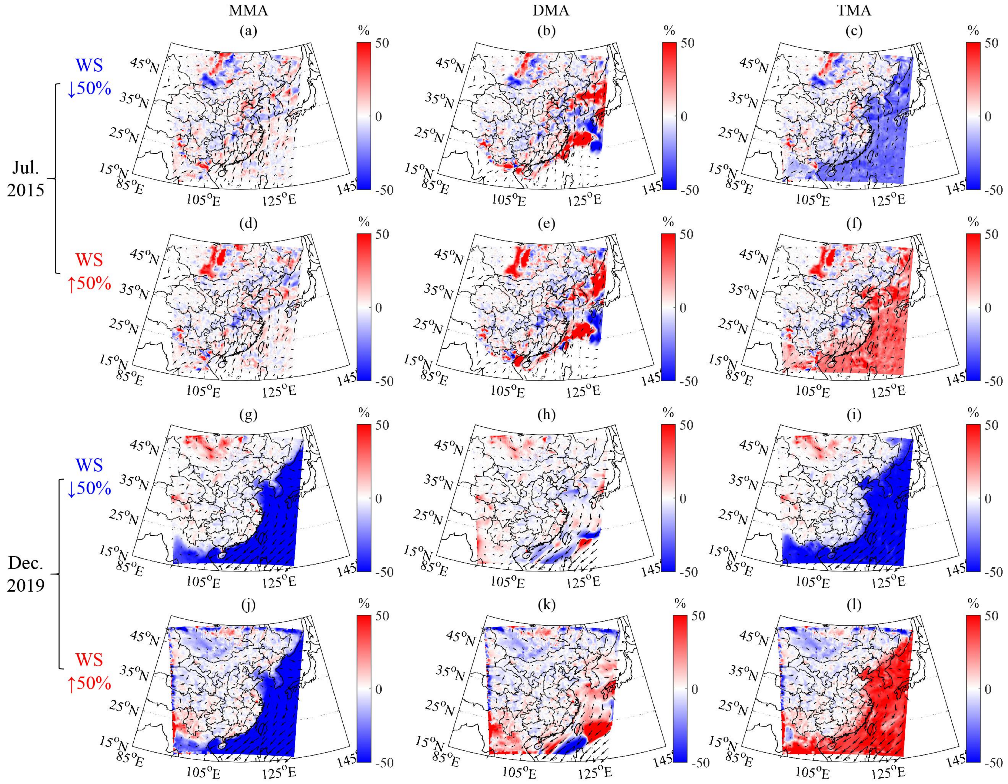


Figure S3: Changes in amines simulated concentrations after the 50% reduction and 50% increase in Chla in WS.

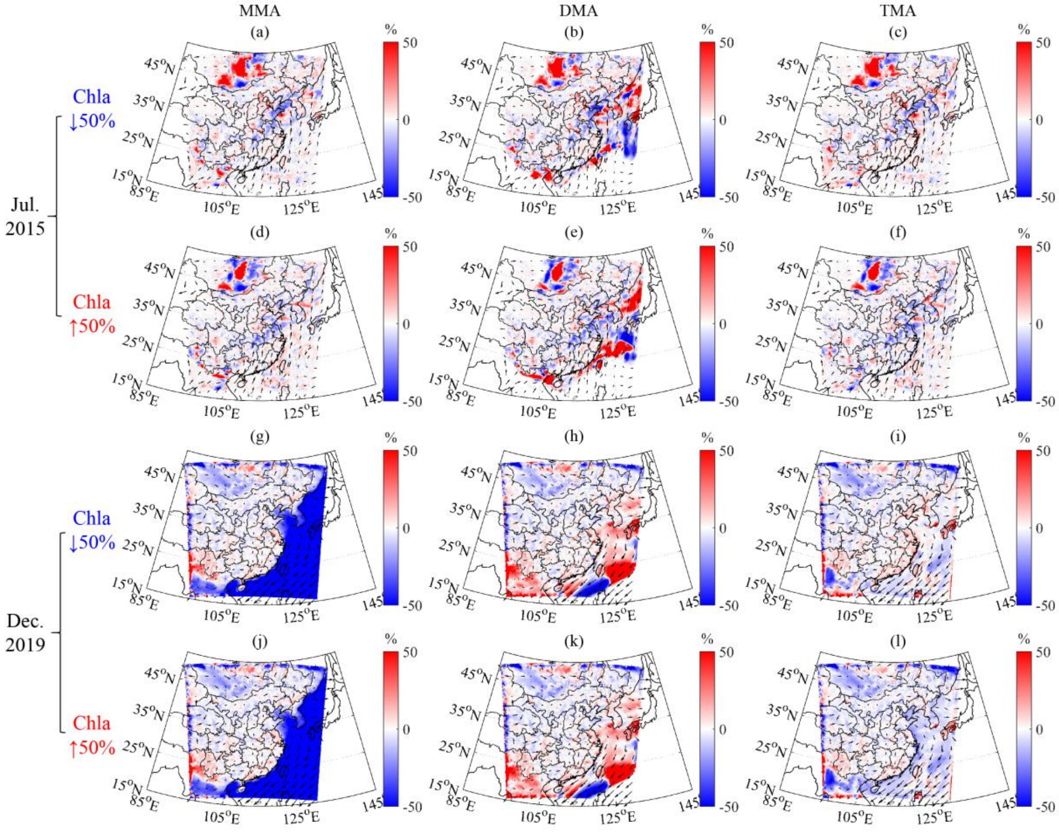


Figure S4: Changes in amines simulated concentrations after the 50% reduction and 50% increase in Chla.

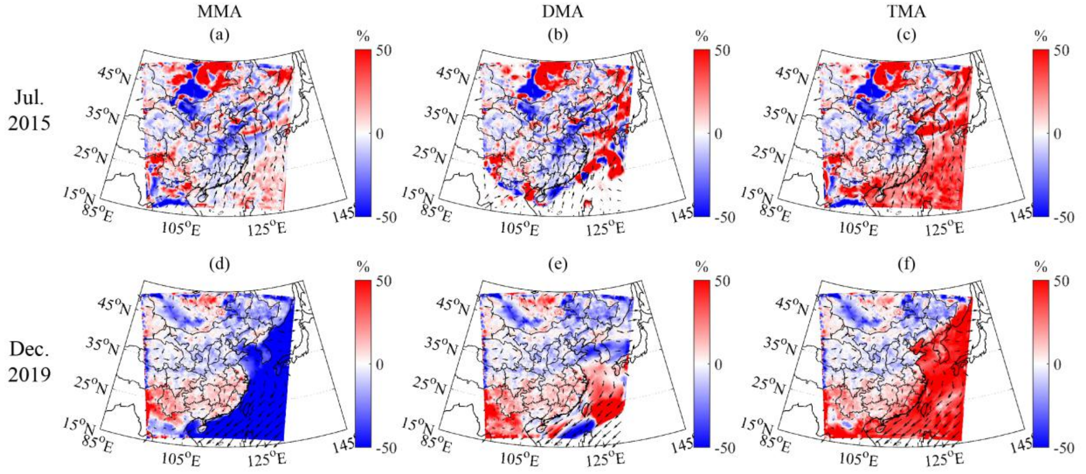


Figure S5: Changes in amines simulated concentrations after the 50% increase in $[C^{+}_{(s)tot}]$.

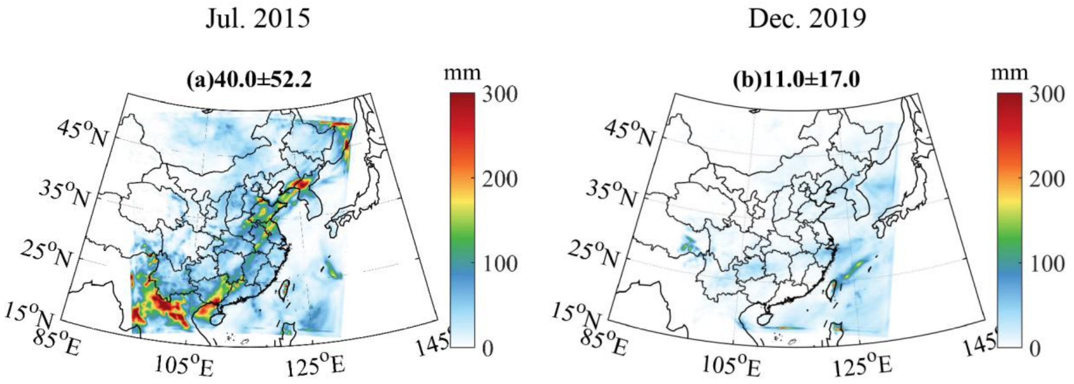


Figure S6: Simulated accumulated rainfall distribution in the simulated period: (a)July 2015, and (d) December 2019.

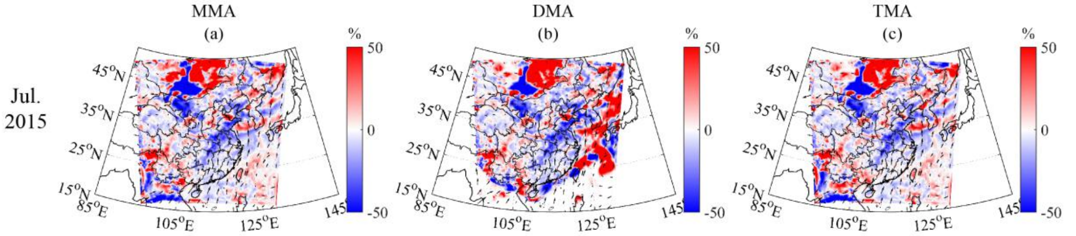


Figure S7: Changes in amines simulated concentration after increasing the apparent Henry coefficient.

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