Reply to Interactive comment on "Hydroxymethanesulfonate (HMS) Formation under Urban and Marine Atmosphere: role of aerosol ionic strength" by Rongshuang Xu et al.

Anonymous Referee #2

The manuscript presents a detailed study of HMS formation in urban and marine environments, examining the occurrence of a haze event in urban Nanjing. An ion chromatography method is used to analyze collected samples for HMS quantification and ISORROPIA II and AIOMFAC were used to estimate ALWQ, pH and ionic strength (IS). An interesting effect of the ionic strength in HMS formation is discussed, providing evidence of HMS enhancement at low ionic strength during high humidity and pollution days. Although the study is well structured it lacks support of some key results, raising questions regarding the pH estimation, and model results vs measurements. The results are interesting, and overall, the work is suitable for publication in "Atmospheric Chemistry and Physics (ACP)" provided that the comments outlined below are adequately addressed.

We thank the reviewer for his/her thoughtful comments. The referee's comments are below in italics followed by our responses in normal font.

Comment #1:

The authors use hydrogen peroxide to react the available S(IV) species, since the ion chromatography signal presents HMS as sulfite. Although hydrogen peroxide is not expected to decompose HMS at pH<6, have the authors tested this reaction for the examined conditions? Since filter aliquots are used the pH needs to monitored and test samples need to be examined for potential HMS decomposition (even from formed radicals) in the analyzed sample.

Response: Thanks for the advice. We have tested the pH of filter aliquots of two hazy days (December 28th and 31st, 2023) and one clean day (December 24th, 2024) using a pH-meter (Sartorius, PB-10). These aliquots were extracted following the same procedure as described in the **Section S1** (Supporting Information) that a 16 mm punch of each filter was cut and extracted using 10 mL of Millipore water (18.2 MΩ). The selection of these specific days was predicated on their comparatively elevated aerosol pH levels. The determined pH values were 5.68, 5.71, and 5.79, respectively, all falling below the threshold of 6. Besides, half of these aliquots were immediately injected to IC and the other half was injected after 3 hours. The results shown there was less than 5% of difference in HMS level between these IC analyses. Therefore, here we believe the potential decomposition of HMS could be negligible. Following information was added into the **Section S1** in Supporting Information.

Page 10, Line 35: "Considering that HMS is quite stable under acidic conditions (pH<6) but become unstable under alkaline environments and dissociates rapidly into SO_3^{2-}

and HCHO (Seinfeld and Pandis, 2016), here we also tested the pH of filter aliquots of two hazy days (December 28th and 31st, 2023) and one clean day (December 24th, 2024) using a pH-meter (Sartorius, PB-10). The determined pH values were 5.68, 5.71, and 5.79, respectively, all falling below the threshold of 6. And half of these aliquots were immediately injected to IC and the other half was injected after 3 hours. The results shown there was less than 5% of difference in HMS level between these IC analyses."

Comment #2:

The study focuses mainly on aerosol HMS presence, without providing evidence that the HMS formation occurred in the aerosol vs the cloud/fog water phase. Since 66% of HMS is formed in cloud and fog water, an analysis and/or discussion on the fate and formation of HMS in cloud and fog water is required and a comparison with this studies finding in the aerosol phase is necessary.

Response: Thanks for raising this critical point. We agree with the reviewer that cloud could serve as a significant medium for HMS formation due to the large amount water content (CWC). It is noted that 66% of in-cloud HMS formation was a general value during their observation period from November 26th to December 16th, 2015, Beijing (Wang et al., 2024). However, near-surface aerosol HMS formation (HMSp) was found to be comparable to those formed in cloud/fog water (HMSc) during their Episode 1. Moreover, under certain days (e.g., November 28th to 29th), HMSp levels could exceed HMSc level, which was attributed to that the enhancing effect of IS can counterbalance the constraints imposed by lower aerosol water content and pH values compared to cloud water. However, the exact extent and scope of this compensatory mechanisms remain ambiguous. In addition, another work has reported the detection of HMS in aerosols regardless of the presence of cloud/fog in the winter of Beijing, and HMS concentrations were found to show a good correlation ($R= 0.92$, $P< 0.01$) with aerosol liquid water content (ALWC) (Ma et al., 2020). Therefore, they proposed that aerosol water can serve as a medium for HMS formation despite the fact that HMS levels were significantly lower (< 1 μg m^{-3}) in the absence of cloud/fog and peak HMS level of 18.5 μg m^{-3} was observed during fog processes. Furthermore, previous study has emphasized that although in-cloud formation processes can be much faster compared to aerosol water given its large water content, their contribution to the near-surface aerosol composition can be negligible during winter haze in Northern China due to the accompanied temperature inversions and high atmospheric stability so that the weak vertical exchange impeded the transportation of gas precursors emitted near surface to high altitudes and the chemicals produced in the high-altitude cloud could not be easily transported to the ground (Wang et al., 2022).

In our current work, no prolonged fog events (RH > 90% and Visibility < 1 km) lasting over two hours were observed as shown in **Figure S2e**, which was attached below for the reviewer's reference. Here, we also utilized the vertical temperature profile obtained from http://weather.uwyo.edu/upperair/sounding.html to identify temperature inversion in the atmospheric boundary layer in the winter of Nanjing. As

shown in **Figure S3**, a predominant occurrence of temperature inversions was noted across our observation days, characterized by inversion layers extending from the surface to altitudes of 2000 m, particularly evident on hazy days. Besides, days without temperature inversions (i.e., from December 19th to December 23th, 2023) were associated with negligible cloud water content (**Figure S2f**), which was obtained from MERRA-2 (Modern-Era Retrospective analysis for Research and Applications, Version 2) (Gelaro et al., 2017). In line with Ma et al.'s work (2020), we also detected HMS within $PM_{2.5}$ samples during 7 of non-cloud days. Besides, our data indicated no discernible correlation between CWC and HMS levels whereas HMS levels displayed a notably positive correlation with ALWC ($R=0.67$, $P<0.01$). While we recognize that the HMS formation in cloud layers characterized by persistent dense clouds and extended durations may carry great contributions, **in the context of this study, the presence of temperature inversions, low wind speeds, and diminished cloud water content during the winter season in Nanjing suggests that the impact of in-cloud HMS formation on the observed levels of particulate HMS may be insignificant**. **Section 3.2** has been extensively revised to provide a more detailed explanation of the role of in-cloud formation in observed particulate HMS levels during our winter observations in Nanjing. We have also made edits in the **Abstract**.

Revision in Abstract:

Page 2, Line 1: "Hydroxymethanesulfonate (HMS) has emerged as a critical organosulfur species in ambient aerosols, yet the impact of aerosol properties, particularly ionic strength (IS), on the formation of HMS remains uncertain. Here, HMS levels in wintertime of urban Nanjing, China were quantified at 0.30±0.10 μg m^{-3}, where the contribution of in-cloud formation likely carry minor significance due to the barrier resulted from stable stratification."

Revision in Manuscript:

Page 8, Line 1: "

3.2 Elevated HMS level during a haze pollution period in urban Nanjing

In addition, a seven-day haze pollution event (from December 27th, 2023 to January 2nd, 2024) was observed in urban Nanjing, where a notably elevated levels of HMS and sulfate were observed ($P<0.05$). Throughout the haze event ($PM_{2.5}$ = 114.3±18.0 μg m^{-3}), the average concentrations of HMS and sulfate were 0.36±0.09 and 11.4±4.0 μg m^{-3}, respectively, with HMS/sulfate molar ratio of 3.5±0.7% (**Table S1**). It is noted that even under hazy days, the HMS level in our observations were significantly lower compared to those reported in Northern China during severe winter haze episode (averaged at 4-7 μg m^{-3}) (Ma et al., 2020; Liu et al., 2021; Chen et al., 2022; Wang et al., 2024). Such divergence can be largely attributed to the contribution of HMS formed in cloud/fog processes, as evidenced by a previous study reporting comparable levels of HMS (< 1 μg m^{-3}) in Beijing to this work in the absence of cloud/fog in Beijing in the absence of cloud/fog, while peak HMS levels of 18.5 μg m^{-}

^{3} was observed during fog processes (Ma et al., 2020). During our sampling period in Nanjing, there were no prolonged fog events (RH>90% and Visibility<1 km) lasting over two hours (**Figure S2e**). Additionally, previous study has emphasized that although in-cloud formation processes can be much faster compared to aerosol water given its large water content, their contribution to the near-surface aerosol composition can be negligible during hazy days in Northern China due to the accompanied temperature inversions and high atmospheric stability so that the weak vertical exchange impeded the transportation of gas precursors emitted near surface to high altitudes and the chemicals produced in the high-altitude cloud could not be easily transported to the ground (Wang et al., 2022). Here, we utilized the vertical temperature profiles retrieved from http://weather.uwyo.edu/upperair/sounding.html to identify temperature inversion within atmospheric boundary layer in the winter of Nanjing. As shown in **Figure S3**, a predominant occurrence of temperature inversions was noted across our observation days, characterized by inversion layers extending from the surface to altitudes of 2000 m, particularly evident on hazy days. Furthermore, days lacking temperature inversions (from December 19th to December 23th, 2023) were associated with negligible cloud water content (**Figure S2f)**, which were obtained from MERRA-2 (Modern-Era Retrospective analysis for Research and Applications, Version 2) (Gelaro et al., 2017). In line with Ma et al.'s work (2020), we also detected HMS within $PM_{2.5}$ samples during 7 of non-cloud days. Therefore, given the protective effects of temperature inversion and low wind speeds (**Figure S2a**) as well as reduced cloud water content prevalent during our observations, the contribution of in-cloud/fog HMS formation to observed particulate HMS levels was likely insignificant and the notably lower HMS level further suggest that the aerosol bulk water might serve as a predominant medium for HMS formation during our observations.

Other factors such as the level of precursors and atmospheric oxidants may also contribute to this divergence. For instance, during the winter haze in Beijing, the average level of HMS, sulfate were reported to be 4 μg m^{-3} and 45 μg m^{-3}, respectively, with HMS/sulfate of 10%, and the gaseous SO_2 and HCHO concentrations were recorded at 12 and 20 ppb, respectively (Ma et al., 2020). This study observed significantly lower levels of SO_2 (2.2±0.7 ppb) and HCHO (5.4±1.1 ppb) during the haze event. On the other hand, the reduced HMS/sulfate ratio could potentially be elucidated by the higher O_3 level in this work (43.3±4.7 ppb) compared to Beijing (5 ppb) as low atmospheric oxidation level could encourage more SO_2 to participate in the formation of HMS rather than sulfate (Song et al., 2019; Ma et al., 2020; Campbell et al., 2022)."

Revision in Supporting Information: "

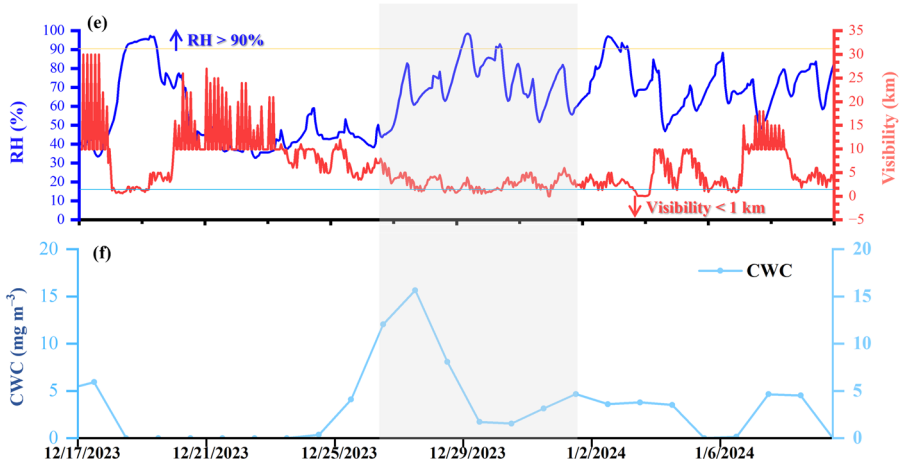

Figure S2. Ancillary atmospheric measurements in urban Nanjing including fog event criteria based on hourly relative humidity (RH>90%) and Visibility (<1 km) data (e); Time series of the average cloud water content below the planetary boundary layer height over our observation sites (f).

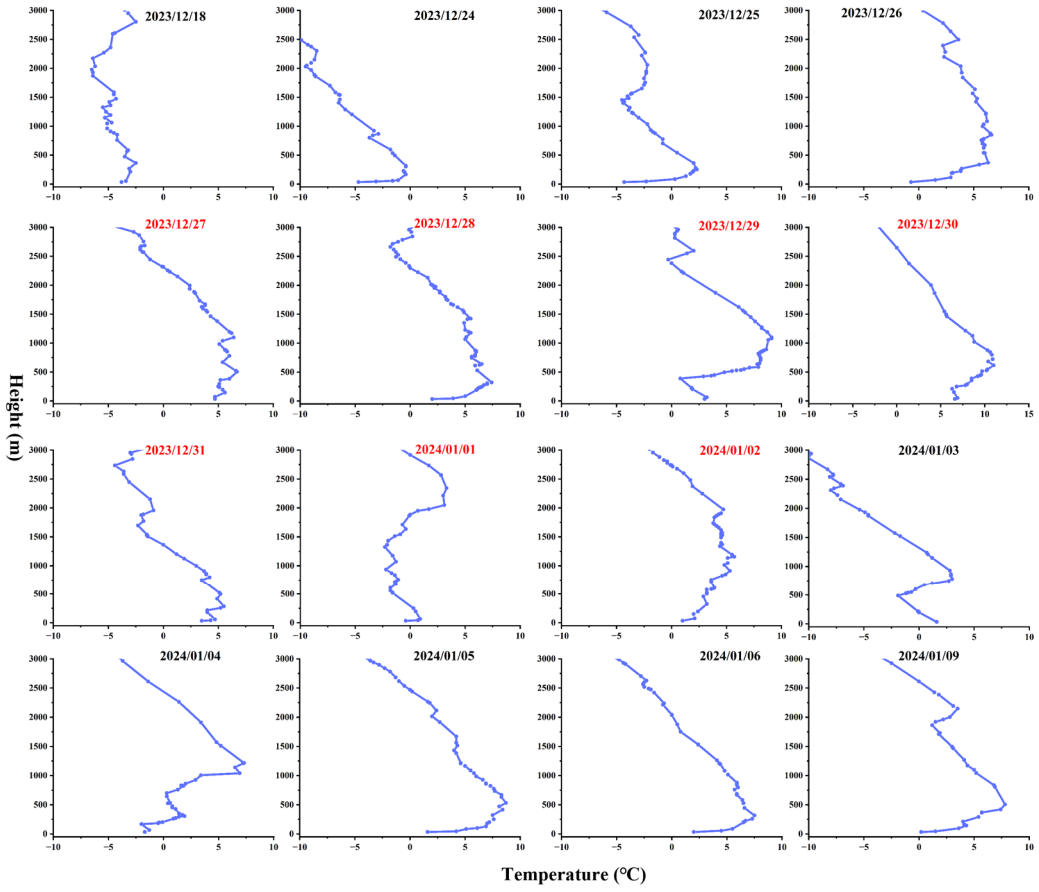

Figure S3. Observed vertical temperature profile at 00 UTC during our observation in Nanjing. The dates of hazy days were marked by red.

As for marine environment, we attempt to determine the HMS formation rate in cloud water ($P_{HMS,\,c}$) and aerosol water ($P_{HMS,a}$) considering the minimal impact of temperature inversion during our observation in April. In the manuscript, we have compared the $P_{HMS,\,a}$ (2.06×10^{-5} μg m^{-3} h^{-1}) in our study with $P_{HMS,\,c}$ (1.2×10^{-5} μg m^{-3} h^{-1}) estimated for observations near Beaufort Sea with a CWC of 4.8 mg m^{-3} and a pH of 5 (Liu et al., 2021). Using these same CWC and pH, here we calculated the averaged $P_{HMS,c}$ of 4.95×10^{-5} μg m^{-3} h^{-1} in our marine environment, which was comparable to the $P_{HMS,\,a}$. The calculation results and input parameters were summarized in **Table R1**. This comparable $P_{HMS,a}$ and $P_{HMS,c}$ can be explained by that despite that the CWC being ~ 300 times higher than ALWC and aerosol pH being lower, the higher ionic strength level in marine aerosol water can significantly promote HMS formation process, rendering the HMS formation within aerosol water a process comparable to that observed in cloud and fog environments. Altogether, although these formation rates serve as initial approximations, **these results may indicate the significant contribution of both aerosol and cloud water in the formation of HMS in marine atmosphere**. Following discussion has been included in **Section 3.4**.

Page 17, Line 10: "Using the averaged [$SO_{2(g)}$]=0.82 ppb and [$HCHO_{(g)}$]=0.5 ppb (Ervens et al., 2003; Zhao et al., 2024), the potential HMS formation rates in these marine aerosols were calculated to be 2.06×10^{-5} μg m^{-3} h^{-1}, a value comparable to reported cloud/fog-based HMS production rate (1.2×10^{-5} μg m^{-3} h^{-1}) near Beaufort Sea (Liu et al., 2021) with a liquid water content (LWC) of 4.8 mg m^{-3} and a pH of 5. Considering the minimal impact of temperature inversion during our observation in April, here we also determined the in-cloud HMS formation rate to be 4.95×10^{-5} μg m^{-3} h^{-1} in our marine environment using a LWC of 4.8 mg m^{-3} and a pH of 5. These results further highlighted the significance of IS-dependent enhancement in HMS formation, which can render the HMS formation within aerosol water a process comparable to that observed in cloud and fog environments, particularly in humid and pristine conditions."

Besides, for the reviewer's reference, here we also attempt to compared the HMS formation rate in cloud water ($P_{HMS,\,c}$) and aerosol water ($P_{HMS,\,a}$) in urban Nanjing although the transportation of in-cloud HMS formation can be largely impeded. Global model (Song et al., 2019) has estimated the cloud pH in the winter of Nanjing aera to be around 4-5, consistent with winter observations in China (4.1±0.6) (Shah et al., 2020). In our analysis, we utilized the upper value of cloud pH of 5 to estimate $P_{HMS,\,c}$ under pre-haze and haze conditions, comparing it with $P_{HMS,\,a}$. Despite the tendency for cloud pH to decrease during haze pollution (Li et al., 2017), it was observed that $P_{HMS,\,c}$ was five times lower than $P_{HMS,\,a}$ on hazy days (**Table R1**). Nevertheless, it is worth noting that the $P_{HMS,\,a}$ value of $1.06\pm0.64 \times 10^{-2}$ μg m^{-3} h^{-1} can effectively reflect the HMS levels observed in ambient aerosol, furthering indicate the predominate role of aerosol water for HMS formation during temperature inversion. Besides, the $P_{HMS,\,c}$ were found to be comparable to $P_{HMS,\,a}$ on clean days. Considering possible overestimations in $P_{HMS,\,c}$ due to reduced precursor concentrations and hindered transport to ground level resulting from weak vertical exchange, these results suggest

the limited impact of in-cloud HMS formation on detected HMS levels. We acknowledge that uncertainties may exist in the calculation of HMS formation rate due to potential variability associated with cloud pH, thus above information was not included in the Manuscript.

Table R1. Summary of $P_{HMS,\,a}$ and $P_{HMS,\,a}$ as well as input parameters.

	Marine		Urban			
	Cloud	**Aerosol**	**Cloud**		**Aerosol**	
			Pre-haze	Haze	Pre-haze	Haze
T (K)	283±1	283±1	272±2	277±4	272±2	277±4
SO_2 (ppb)	0.82±0.42[a]	0.82±0.42	1.99±0.45	2.21±0.69	1.99±0.45	2.21±0.69
HCHO (ppb)	0.5 [a]	0.5	1.91±1.05	5.38 ±1.08	1.91±1.05	5.38 ±1.08
LWC ($\mu g\ m^{-3}$)	4.8 [b] mg m^{-3}	14.72	1.5±2.2 mg m^{-3}	5.12±5.45 mg m^{-3}	22.69±20.65	80.45±38.40
pH	4.8 [b]	4.24±0.15	5 [d]	5	5.25±0.42	4.76±0.46
Ionic strength (mol kg^{-1})	10^{-4} [c]	4.30±1.24	10^{-4}	10^{-4}	12.32±3.19	8.85±1.30
P_{HMS} ($\times 10^{-4}$ μg $m^{-3}\ h^{-1}$)	0.51	0.21	2.56±4.68	20.4±23.3	6.40±6.96	106±64.6

[a] The levels of SO_2 and HCHO over marine environments were adopted from previous studies (Ervens et al., 2003; Zhao et al., 2024).
[b] The LWC and pH for cloud/fog droplets came from previous coastal study near Beaufort Sea (Liu et al., 2021).
[c] This typical ionic strength value of cloud droplet was given by Herrmann et al. (2015).
[d] This cloud pH was the upper value of global estimation on Nanjing aera (Song et al., 2019).

Comment #3:

The estimation of pH is achieved without ammonia measurements. How accurate is this estimation? More information is needed.

Response: Thanks for the comment. To clarify, we monitored the levels of gas-phase ammonia during our observations in Nanjing using a Monitor for AeRosols and GAses (MARGA; Metrohm Ltd., Switzerland) to estimate urban aerosol pH as detailed in **Section 2.1** (Page 5, Line 15). The daily averaged NH_3 levels were added into Supporting Information as **Figure S2c** (attached below). For marine aerosol, we utilized the $\varepsilon(=NH_4^+/(NH_3+NH_4^+))$ value from previous study which conducted their observations in the Bohai Sea in May 2021 (Wang et al., 2022), where the NH_3 concertation averaged at 3.4±1.6 and the NH_4^+ level averaged at 4.0±3.7 μg m^{-3}. Considering the resemblance of our observational environment in April 2023 in Bohai Sea and Huanghai Sea to that of the referenced study, we used a ε value of 0.5 to determine the marine aerosol pH. We would like to declared that there was an inadvertent citation error in our previous reference and we have corrected this in the Manuscript.

Page 6, Line 3: "For marine aerosols, we adopted a particle to particle + gas partitioning fraction of NH_4^+ (ε) of 0.5, drawing from prior observations conducted in the Bohai Sea in May (Wang et al., 2022) in light of the absence of gaseous data."

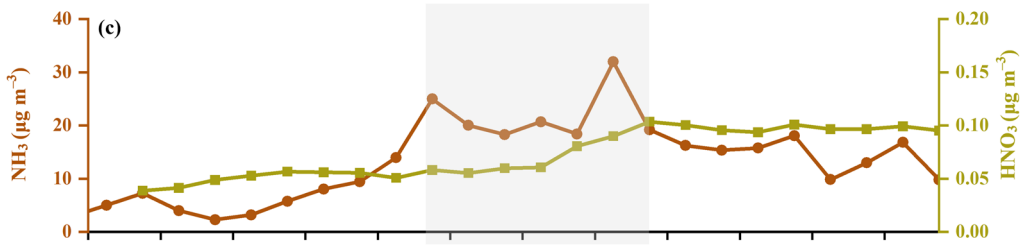

Figure S2. Ancillary atmospheric measurements in urban Nanjing including gas-phase NH_3 and HNO_3 level (c).

Comment #4:

The model results are interesting, providing new insights on HMS fate. However, with the exception of correlations, how do these results compare with the measurements? What are the most sensitive parameters of the model and what is the deviation between measurements and model results. How does this affect the conclusions?

Response: Thanks for the comment. As shown in **Figure 2** (attached below), this work estimated larger HMS formation rates (P_{HMS}) during haze events compared to clean days (**Figure 2f**), showcasing a consistent trend with ambient HMS level measurements (**Figure 2c**). In addition, considering other atmospheric process such as physical transport could also impact ambient HMS level, we has utilized the HMS to carbon dioxide (CO) ratio to better represent the secondary formation of ambient HMS as CO was usually considered to be an inert chemical species during rapid haze formation, with its variation often interpreted as indicative of the accumulation of primary pollutants in the shallower boundary layer (Williams et al., 2016). Therefore, the larger HMS/CO ratio observed during haze events further evidenced the enhanced HMS formation, in line with our P_{HMS} results. The P_{HMS} estimations also exhibited a good correlation with HMS/CO ($R=0.57$, $P<0.05$), and can roughly capture the diurnal variations of HMS/CO during pollution period (**Figure S7a**, **attached below**).

Regarding the comparison between predicted P_{HMS} and HMS measurements, a previous study attempted to correlate P_{HMS} (μg m^{-3} h^{-1}) and daily averaged HMS levels by multiplying 24 hours with P_{HMS} as a rough approximation (Shen et al., 2024) given the low reactivity of HMS after its formation although other physical process such as diffusion and deposition can also impact its ambient concentration (Liu et al., 2024). As shown in **Figure R1**, our comparison between predicted HMS and measured HMS levels exhibited a consistent pattern, with higher concentrations during haze events. The averaged HMS level predictions during hazy days were in line with ambient observations, whereas the estimated HMS level was an order of magnitude lower than

the measurements on clean days. This discrepancy can be largely attributed to the fact that ambient HMS levels are influenced by various key atmospheric processes, including precursors emission, physical transport (advection and turbulent diffusion), chemical reactions, and deposition losses, whereas P_{HMS} solely represents the averaged rate for the HMS formation process. While we acknowledge that chemical transport models offer a more comprehensive representation of these atmospheric processes and can provide more precise estimations of HMS levels, the primary focus of our P_{HMS} calculations was to emphasize the enhanced HMS formation during hazy days, as evidenced by the higher HMS/CO ratio, and the significance of aerosol ionic strength in this context. Despite potential uncertainties may persist in P_{HMS} calculations reported here due to the lack of laboratory-based kinetics under higher ionic strength level (i.e., IS > 11 mol kg^{-1}) and potential enhancement of HCHO solubility within aerosol water as detailed in the Manuscript (Page 13, Line 35), **these P_{HMS} calculations were intended as an initial approximation to depict the potential fluctuations of HMS formation rates under different pollution conditions, rather than for direct comparison with ambient HMS levels.**

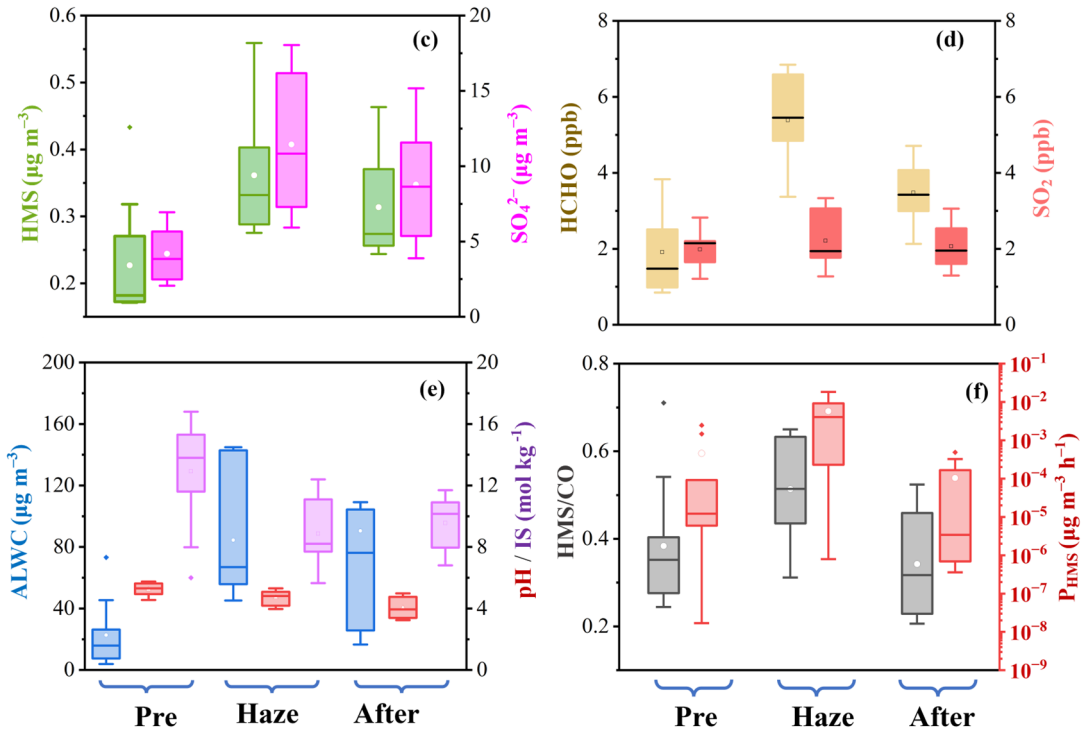

Figure 2. The averaged atmospheric characteristics under different pollution scenarios in urban Nanjing (i.e., pre-haze, haze and after) including: HMS and sulfate concentration (c); gas precursors levels (d); aerosol properties such as liquid water content (ALWC), pH and ionic strength (IS) (e); and comparison (f) between HMS/CO ratio and HMS formation rates (P_{HMS}).

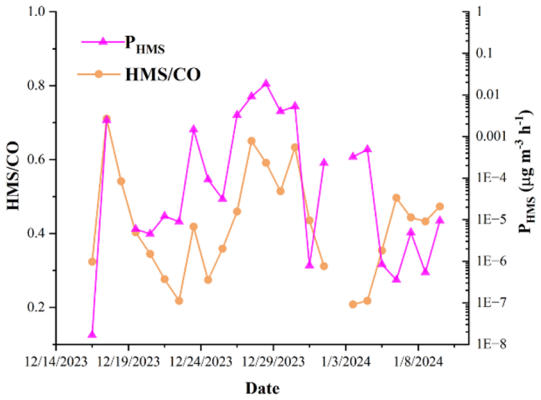

Figure S7a. The comparison between HMS formation rates (P_{HMS}) and HMS/CO ratio in urban Nanjing.

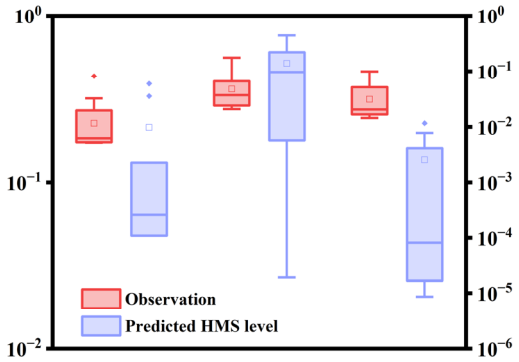

Figure R1. Comparisons between observed HMS level and predictions using HMS formation rates.

Comment #5:

The HMS to sulfate ratio is mass or molar ratio?

Response: Thanks for the hint. The ratio is molar ratio. We have specified it in the Manuscript and Supporting Information.

Comment #6:

Page 6: Why is an epsilon of 5 for NH3/NH4+ partitioning used for the present study? It is stated that is based on Fairbanks Alaska, but this works investigated environment is significantly different and with higher pollution.

Response: Thanks for the comment. To clarify, we monitored the levels of gas-phase

ammonia during our observations in Nanjing using MARGA to estimate the urban aerosol pH. For marine aerosol, we utilized the ε(=NH_4^+/(NH_3+NH_4^+)) value based on previous observations conducted in the Bohai Sea in May 2021 (Wang et al., 2022), where the NH_3 concertation averaged at 3.4±1.6 and the NH_4^+ level averaged at 4.0±3.7 μg m^{-3}. Considering the resemblance of our observational environment in April 2023 to that of the aforementioned study, here we used a ε of 0.5 to estimate the marine aerosol pH. We would like to declared that there was an inadvertent citation error in our previous reference and we have corrected this in the Manuscript.

Page 6, Line 2: "For marine aerosols, we adopted a particle to particle + gas partitioning fraction of NH_4^+ (ε) of 0.5, drawing from prior observations conducted in the Bohai Sea in May (Wang et al., 2022) in light of the absence of gaseous data."

Comment #7:

Page 8 Lines 18-21: This statement is a bit confusing. Since HCHO is a precursor of HMS, wouldn't it be expected to observe a decrease of HCHO and an increase of HMS rather than simultaneous peaks of the two species?

Response: Thanks for the comment. We agree with the reviewer that the formation of HMS entails the consumption of its precursor formaldehyde (HCHO). Nevertheless, given the minimal presence of HMS (specifically, HMS level at 0.36±0.09 μg m^{-3} and HCHO level at 6.72±1.56 μg m^{-3} during hazy days), the generation of HMS likely exerts a negligible influence on ambient HCHO concentrations. In this scenario, it was the level of HCHO that predominantly influences the concentration of HMS.

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