Response to reviewer's comments

Reviewer #1

We would like to thank the authors for providing a comprehensive observational dataset of particulate amines over the Yellow Sea–Bohai Sea region. Filter-based measurements of low molecular weight amines in marine and coastal aerosols remain scarce, and the presented dataset adds valuable information on amine concentrations and speciation in this region, particularly in relation to co-measured inorganic and organic aerosol components.

At the same time, we suggest that the manuscript may be better suited for publication as a **measurement report**, with the primary emphasis placed on the observational dataset and its descriptive analysis. While the interpretations and proposed formation pathways are generally plausible and well-motivated by previous literature, many of the conclusions rely on correlation-based approaches and remain qualitative. Framing the study as a measurement report would highlight the clear strength of the work, especially the quality and breadth of the observations, while avoiding over-interpretation of secondary formation mechanisms and source contributions that cannot be constrained by the present dataset. Therefore, we suggest revising the manuscript title to reflect the measurement report, highlighting the detection of low molecular weight amines.

This review reflects a **joint assessment by two independent reviewers,** and we have discussed the manuscript together. The comments below, therefore, represent a consolidated perspective of the submitted preprint.

Sincerely thanks for the reviewer's comment! We have read carefully, revised and responded to each point. **The modified contents are shown in bold font.**

Author's response:

We sincerely thank the reviewers for this insightful suggestion. We agree that our dataset provides a comprehensive observational basis, which is indeed a key strength of this study. However, we respectfully consider that the manuscript is more appropriately framed as a Research Article rather than a measurement report.

Beyond presenting observational data, this work attempts to advance the understanding of the sources and formation mechanisms of low molecular weight amines in marine aerosols. (1) We identified distinct compositions of amines in offshore aerosols compared to terrestrial aerosols, with TMDEA surpassing MA as the predominant amine. Nevertheless, MA remained the second most abundant amine, particularly under the influence of terrestrial air masses. This has not been fully concerned in previous studies, particularly in the YS−BS region. (2) We provided new observational constraints on the sources and atmospheric processing of amines in marine aerosols using specific organic molecular tracers representing primary biogenic sources, higher plant waxes, marine/microbial sources, biogenic secondary organic aerosols, biomass burning, and fossil fuel combustion. This approach has not been applied in previous studies. Results suggested that individual amines were associated with different primary sources (terrestrial vs. marine; biogenic vs. anthropogenic), and two distinct major secondary formation pathways (nitrate vs. sulfate associated) were inferred. (3) Our findings provided new insights in continental and marine influences of amines in offshore aerosols in the context of existing literature. Terrestrial sources not only emit gaseous amines but also contribute acidic aerosols that can further uptake amines from marine sources during the transportation of air masses from the mainland to the ocean. The secondary formation of aerosol amines were influenced by both precursors abundance and ambient conditions. The correlations between aerosol amines and NO_3^- or SO_4^{2-} cannot be regarded as the sole evidence for determining the contribution from anthropogenic sources of amines.

We acknowledge that many of the conclusions are based on correlation analyses and remain qualitative, however, we have made every effort given the inherent limitations in data availability and the difficulty of constraining sources for marine aerosols. To address the reviewer's concern regarding potential over-interpretation, we have revised the manuscript to further clarify the limitations of our dataset and temper the language where appropriate, particularly in the discussion of secondary formation pathways and source contributions. We ensured that all conclusions are presented more cautiously and are clearly supported by the available evidence.

Given these considerations, we hope the reviewers will agree that the scope and contributions of this work justify its classification as a Research Article.

General comments:

The authors present total suspended particle (TSP) filter-based measurements collected during a Chinese oceanographic cruise over the Yellow Sea and Bohai Sea (BS). Chemical analysis using ion chromatography was performed to quantify six major protonated amine species and several other organic and inorganic ions.

Particle-phase amines have been reported in the Chinese coastal marine atmosphere due to their role in secondary aerosol formation. Considering the breadth of existing research on particulate amines, particularly in the Chinese coastal environment, the author should clearly highlight the novelty and added value of this work. Given that the measurements were conducted in 2018, the authors should clearly articulate how this dataset advances existing knowledge beyond what is already known.

Author's response: Thanks for the comment.

We have highlighted the novelty and added value of this work in Introduction (Lines 117-137) and Conclusions (Lines 553-577). Although the measurements were conducted in 2018, we believe that our approach and results advance existing knowledge and provide useful insights for future studies.

"**Previous studies on aerosol amines over the marginal seas of China have mainly focused on DMA and TMDEA, the sum of TMA and DEA (Zhou et al., 2019; Xie et al., 2018; Yu et al., 2016; Hu et al., 2015). Although MA has been observed as the dominant amine in urban aerosols in northern China and the Yangtze River Delta region (Yang et al., 2023; Liu et al., 2023; Huang et al., 2018), its contribution in marine aerosols of China remains unclear. The primary sources and secondary formation pathways of aerosol amines over the YS−BS are poorly constrained due to the combined influence of complex terrestrial and marine emissions, as well as the lack of specific source indicators.** To address these, an integrated analysis of six major amines together with more than 100 other chemical components in aerosols was conducted using filter samples collected over the YS−BS during a research cruise in spring 2018. Spatial variations, potential sources, and secondary formation pathways of aerosol amines were investigated. **By elucidating the relationships between individual amines and specific organic molecular tracers representing six source categories, this study provides new observational constraints on the sources and atmospheric processing of amines in marine aerosols. The results suggest that individual amines were associated with different primary sources and likely underwent two distinct major secondary formation pathways. These findings provide a basis for improving the quantitative source apportionment of aerosol amines and for further clarify their origins and gas-to-particle conversion under varying ambient conditions.**" (Lines 117-137)

"**Offshore aerosols exhibited distinct compositions of amines compared to terrestrial aerosols, with TMDEA surpassing MA as the predominant amine.** The proportions of TMDEA in ∑amines and the relative contributions of ∑amines in aerosols increased from north to south (BS < NYS < SYS), highlighting the ocean as a substantial source of amines, particularly TMDEA, despite the significant influence of terrestrial emissions. **Distinct potential sources and major secondary formation pathways were identified for different amine species.** MA, EA, and DMA were mainly derived from terrestrial biogenic and non-combustion anthropogenic sources, followed by fossil fuel combustion, with over 50% formed via nitrate-associated secondary formation pathways, interacting with BSOA formation in the NO_x-involved oxidation of BVOCs. In comparison, PA mainly originated from combustion-related sources along with terrestrial and marine biogenic sources, with only ~35% contributed by nitrate-associated secondary formation. In contrast to other amines, TMDEA was mostly (~60%) generated via sulfate-associated secondary formation pathways, and also contributed by primary marine aerosols from sea spray and bubble bursting.

Terrestrial sources not only emit gaseous amines but also contribute acidic **aerosols that can further uptake amines from marine sources during the transportation of air masses from the mainland to the ocean. This process affects the physiochemical properties and climate effects of marine aerosols, as well as the carbon and nitrogen cycles. In addition to precursors abundance, ambient conditions also influence the secondary formation of aerosol amines, leading to temporal and spatial variations in their concentrations and compositions. Overall, these findings improve the understanding of amines in marine aerosols, highlight the impact of terrestrial emissions on offshore aerosol chemistry, and underscore the importance of multiphase chemical processes of amines under diverse ambient conditions.**” (Lines 553-577)

There are numerous studies focused on the coastal/marine regions of China. While the authors have cited many of those, they have missed a few relevant studies (e.g., Chen et al., 2022; Huang et al., 2018; Du et al., 2021). In addition, a recent perspective review article by Kanawade and Jokinen (2025), which discusses challenges in detecting minute levels of amines in both gas- and particle-phases and emphasises the importance of such measurements, would be highly relevant.

Author’s response: Thanks for the comment.

We have added these citations as suggested.

Lines 19-22: Amines and other chemical components were not directly detected or measured in the air; rather, they were extracted from aerosol samples collected on filters. This distinction should be clearly stated throughout.

Author's response: Thanks for the comment.

We have revised as suggested (Lines 18-23), and descripted more clearly throughout.

"Here, an integrated observation of methylamine (MA), ethylamine (EA), dimethylamine (DMA), iso-propanamine (IPA), propanamine (PA), "trimethylamine + diethylamine" (TMDEA), and over 100 other chemical components was conducted **in total suspended particles samples collected** during a spring 2018 research cruise across the Yellow Sea and Bohai Sea, China." (Lines 18-23)

Line 72: Myriokefalitakis et al. assume that amines account for approximately one-tenth of the ocean-derived ammonia; however, this contribution may be up to three orders of magnitude lower in reality, which would substantially reduce the estimated role of amines in marine SOA formation. This limitation should be stated more clearly.

Author's response: Thanks for the comment.

We have added the limitation (Lines 75-80).

"Global modeling (Myriokefalitakis et al., 2010) suggested that amines contribute ~20% of marine SOA, ranking second to dimethylsulfide (DMS). **However, this contribution may be substantially overestimated, given that the actual proportions of amines relative to NH_3 are up to three orders of magnitude lower than the values assumed in the model.**" (Lines 75-80)

Line 120-125: These statements are very. They should be replaced with specific scientific questions or hypotheses that clarify the study's added value to our understanding of amines in Chinese coastal regions, particularly in the YS-BS region.

Author's response: Thanks for the comment.

We have replaced them with specific scientific questions, and clarified our study's added values (Lines 117-137).

"**Previous studies on aerosol amines over the marginal seas of China have mainly focused on DMA and TMDEA, the sum of TMA and DEA (Zhou et al., 2019; Xie et al., 2018; Yu et al., 2016; Hu et al., 2015). Although MA has been observed as**

the dominant amine in urban aerosols in northern China and the Yangtze River Delta region (Yang et al., 2023; Liu et al., 2023; Huang et al., 2018), its contribution in marine aerosols of China remains unclear. The primary sources and secondary formation pathways of aerosol amines over the YS−BS are poorly constrained due to the combined influence of complex terrestrial and marine emissions, as well as the lack of specific source indicators. To address these, an integrated analysis of six major amines together with more than 100 other chemical components in aerosols was conducted using filter samples collected over the YS−BS during a research cruise in spring 2018. Spatial variations, potential sources, and secondary formation pathways of aerosol amines were investigated. **By elucidating the relationships between individual amines and specific organic molecular tracers representing six source categories, this study provides new observational constraints on the sources and atmospheric processing of amines in marine aerosols. The results suggest that individual amines were associated with different primary sources and likely underwent two distinct major secondary formation pathways. These findings provide a basis for improving the quantitative source apportionment of aerosol amines and for further clarify their origins and gas-to-particle conversion under varying ambient conditions.**" (Lines 117-137)

Line 187-194: TMDEA concentrations in TSP are compared with values from earlier studies that reported TMDEA either in PM10, PM2.5 or PM1. Further, most of these studies represent different seasons and sampling periods. While the reported concentrations appear broadly comparable, one-to-one quantitative comparisons are not appropriate. Grouping previous studies by region (e.g., SYS, NYS, BS) may be more informative, given the clear spatial variability of amine and other species. The authors should also comment on concentration differences arising from particle size cut-offs (TSP versus PMn). In my opinion, Section 3.1 should focus primarily on studies conducted in the YS-BS region during spring (April), with only brief reference to other regions or seasons.

Author's response: Thanks for the comment.

We have revised as suggested.

(1) A comment on concentration differences arising from particle size cut-offs has been added.

(2) The discussion of other regions and seasons has been revised and simplified (Lines 204-212).

"The concentrations of amines measured in TSP were comparable to $PM_{2.5}$ and PM_{10} (Table S3), as **amines are predominantly (> 70%) distributed in aerosols with diameters < 1.8 µm** (Zhou et al., 2019; Xie et al., 2018; Yu et al., 2016). **Compared with other marine and coastal regions, the aerosol TMDEA concentrations in spring over the YS−BS were higher than those reported for the East China Sea (ECS), Huaniao Island (in the ECS), South China Sea (SCS), and Northwest Pacific Ocean (NWPO) (Chen et al., 2022; Zhou et al., 2019; Xie et al., 2018; Yu et al., 2016). Over the YS−BS, aerosol TMDEA concentrations were higher in summer than in spring and autumn (Xie et al., 2018; Yu et al., 2016)**." (Lines 204-212)

(3) Other discussions in Section 3.1 have also been revised accordingly.

Amines exhibit a north-to-south decrease, and TMDEA is the predominant amine species in TSP over the YS-BS. However, TMDEA, despite constituting the largest fraction of total amines, does not appear to show a clear north-to-south gradient, whereas MA and EA do. This inconsistency should be addressed.

Author's response: Thanks for the comment.

We have added a linking sentences in Section 3.1 (Lines 242-247), and clarified that this inconsistency can be attributed to differences in sources and secondary formation pathways, which were discussed in detail in Section 3.3.

"**In contrast to MA, EA, DMA, and PA, which exhibited a clear north-to-south decease (SYS < NYS−BS), TMDEA and IPA showed no obvious spatial variation in concentrations. These results suggested that MA, EA, and DMA might share similar sources and secondary formation pathways, whereas IPA, PA, and TMDEA were likely influenced by different sources or atmospheric formation processes.**" (Lines 242-247)

Although the correlations are statistically significant at the 95% (?) confidence level, the number of samples is relatively small. The authors should discuss how the limited sample size may affect the interpretation of the results, particularly given that most analyses rely on a correlation-based approach (Figs. 2,3,4, and S6).

Author's response: Thanks for the comment.

We acknowledge that the number of samples collected during the cruise is limited compared to those from stationary sites, due to the logistical challenges of obtaining marine aerosol samples. Most marine aerosol studies on the South China Sea, East China Sea, Bohai Sea, and Yellow Sea have been conducted during 2-3 week cruise campaigns, typically yielding 10-20 aerosol samples (Yao et al., 2023; Guo et al., 2020; Kang et al., 2018).

For the correlation-based approach, at least 10 observations are generally considered sufficient for preliminary analysis. In this study, the correlation-based approach was employed to provide evidence-based constraints on the potential sources and secondary formation pathways of amines. This approach has also been widely used in previous marine aerosol studies.

We suggest that future studies on marine aerosols could benefit from the use of online measurement techniques or increased sampling frequency, which would better capture spatial and temporal variability and provide stronger constraints on source apportionment.

Additional comments on the interpretation of secondary formation processes:

The manuscript frequently refers to “secondary formation of amines” in the atmosphere. From an atmospheric chemistry perspective, it would be important to clarify more explicitly that low molecular weight amines are predominantly primary emissions in the gas phase, while their occurrence in the particle phase is largely driven by secondary particle-phase formation and partitioning processes (e.g., acid–base reactions, displacement reactions, and dissolution into aerosol liquid water). As currently written, some statements may be interpreted as implying the atmospheric production of new amine molecules, rather than secondary processing of primary gaseous amines.

Author’s response: Thanks for the comment.

We have included a clear definition in Introduction (Lines 49-51), and revised the whole manuscript carefully to avoid misunderstandings.

“These amines are primarily emitted in the gas phase and mainly occur in aerosols as aminium salts formed via **chemically reactive gas-to-particle conversion, commonly referred to as secondary formation of amines.**” (Lines 49-51)

Related to this point, the reported percentage contributions of nitrate- or sulfate-associated secondary formation should be interpreted with caution. These estimates are based on correlation analyses and ion-weighted regression approaches and therefore represent semi-quantitative indicators of association, rather than quantitative source apportionment or mechanistic yields. We recommend that the authors clearly state these limitations and frame the reported percentages accordingly.

Author's response: Thanks for the comment.

We have revised as suggested.

(1) A statement of these limitations has been added.

(2) The weighted standard deviations have been given (Lines 436-442).

"**These estimates are semi-quantitative and limited by the small sample sizes, rather than representing quantitative source apportionment or mechanistic yields.** Contributions of nitrate-associated secondary formation to ∑amines were highest in TSP over the BS (43.0 ± 26.9%), followed by the NYS (33.8 ± 19.7%) and SYS (21.8 ± 18.8%). Among individual amines, nitrate-associated secondary formation contributed most to MA (74.0 ± 61.5%), followed by DMA (65.7 ± 44.3%), EA (52.6 ± 55.0%), and PA (35.1 ± 22.4%)." (Lines 436-442)

(3) Section 3.3.4 have also been revised accordingly.

Several of the proposed formation pathways (e.g. preferential sulfate-driven formation of TMDEA, displacement reactions involving ammonium salts) are chemically feasible and consistent with previous laboratory and theoretical studies. However, given that the dataset is based on filter-based TSP measurements and lacks gas-phase amine data or size-resolved aerosol information, these mechanisms cannot be uniquely constrained by the observations alone. A clearer distinction between observational evidence, inferred associations, and hypothesised mechanisms would strengthen the discussion.

Author's response: Thanks for the comment.

We have improved the expression of whole Section 3.3.3 and Section 3.3.4 to distinguish observational evidence, inferences, and hypothetical mechanism (such as Lines 480-499).

"TMDEA in TSP over the YS−BS showed no correlation with Cl^-, or NO_3^-, but exhibited significant positive linear relationships with SO_4^{2-}, $C_4H_4O_4^{2-}$, and $C_5H_6O_4^{2-}$ (Figure 4 and Figure S6). **The gas-to-particle conversion of TMDEA was inferred to include uptake onto acidic particle surfaces (Equation 2), acid-base reactions with H_2SO_4 (Equation 8) and dicarboxylic acids ($C_4H_6O_4$ and $C_5H_8O_4$), as well as displacement reactions with $(NH_4)HSO_4$ and $(NH_4)_2SO_4$ (Equation 9−10).** Uptake onto acidic particle surfaces is considered as a key step in the gas-to-particle conversion of TMDEA, as TMA exhibits the strongest alkalinity among gaseous amines. TMDEA in TSP over the YS−BS showed limited association with chloride and nitrate, likely due to the much lower competitiveness of TMA in forming these salts (as reflected by dissociation constants) relative to MA, EA, DMA, and NH_3 (Ge et al., 2011). Instead, acid-base reactions with H_2SO_4, together with displacement reactions involving $(NH_4)HSO_4$ and $(NH_4)_2SO_4$, were the major pathways for the secondary formation of aerosol TMDEA. Contributions of dicarboxylic acids were relatively minor, given the significantly lower concentrations of $C_4H_4O_4^{2-}$ and $C_5H_6O_4^{2-}$ compared with SO_4^{2-} in TSP over the YS−BS (Table S2). These findings were consistent with previous laboratory and theoretical studies showing that TMA preferentially reacts with H_2SO_4 (Johnson and Jen, 2023), and that DEA exhibits the highest uptake coefficient during the irreversible reactive uptake of gaseous ethylamines by H_2SO_4 (Yin et al., 2011)." (Lines 480-499)

Finally, while the manuscript discusses implications for new particle formation and aerosol growth, it should be emphasised that the present measurements do not directly probe nucleation or early growth processes, which are typically governed by submicron particles and gas-phase precursors. Any conclusions related to NPF should therefore be framed qualitatively, highlighting implications rather than direct observational evidence.

Author's response: Thanks for the comment.

Considering all reviewers' comments, we have removed all parts related to new particle formation in results and discussion.

Minor comments:

Lines 17: modify as aerosol "physiochemical" properties

Author's response: Thanks for the comment.

We have revised as suggested (Lines 16-17).

"Atmospheric low molecular weight amines play important roles in aerosol **physiochemical** properties and climate." (Lines 16-17)

Line 103: For consistency, replace "the Bohai sea (BS) and Yellow Sea (YS)" with "the Yellow Sea (YS) and Bohai Sea (BS)" throughout.

Author's response: Thanks for the comment.

We have revised as suggested (Lines 109-110).

"The **Yellow Sea (YS) and Bohai Sea (BS)** are two marginal seas in eastern China that serve as transition zones for atmospheric pollutants and particles transported from East Asia to the Northwest Pacific Ocean (NWPO)." (Lines 109-112)

Line 130: Please clarify why a cyclone or any size-selective inlet was not used. Also indicate the sampling duration (e.g., 24 hours?)

Author's response: Thanks for the comment.

(1) Total suspended particulates (TSP) samples were collected instead of using a cyclone or size-selective inlet because our primary objective was to capture the full spectrum of marine airborne particles. This approach allows for a more comprehensive assessment of chemical components of marine aerosols, including both fine and coarse fractions. The particle size distribution of marine aerosols is very different from that of terrestrial aerosols. Primary marine sources such as sea spray and bubble bursting might contribute coarse particles. For the integrity of the source analyze, TSP was selected to collect for analysis. Many marine aerosol studies based on cruises have also collected TSP for investigation (Yao et al., 2023; Wang et al., 2023; Hu et al., 2022; Guo et al., 2020; Kang et al., 2018).

(2) The sampling duration have been added in **Table S1** (see the Appendix).

Line 137: Explain acronyms SYS, NYS

Author's response: Thanks for the comment.

The full name and acronym "South Yellow Sea (SYS)" and "North Yellow Sea (NYS)" have appeared in Introduction (Lines 112-113).

"The YS is divided into **South Yellow Sea (SYS) and North Yellow Sea (NYS)**, both semi-open sea areas of the NWPO." (Lines 112-113)

Line 147: IC systems are used to separate, identify, and quantify compounds in aerosol samples collected on filters, not to measure or detect them directly in ambient air. The terminology should be revised accordingly.

Author's response: Thanks for the comment.

We have revised as suggested (Lines 156-161).

"Six major protonated amine species **extracted from TSP filter samples**, including methylamine ($CH_3NH_3^+$, MA), ethylamine ($CH_3CH_2NH_3^+$, EA), dimethylamine [$(CH_3)_2NH_2^+$, DMA], iso-propanamine [$(CH_3)_2CHNH_3^+$, IPA], propanamine ($CH_3CH_2CH_2NH_3^+$, PA), and the combined species "trimethylamine [$(CH_3)_3NH^+$, TMA] + diethylamine [$(CH_3CH_2)_2NH_2^+$, DEA]" (TMDEA), were measured by a Thermo Fisher Scientific Dionex ICS-5000+ system" (Lines 156-161)

The terminologies "MA", "EA", "DMA", etc. were adopted instead of "$MMAH^+$", "$MEAH^+$", "$DMAH^+$", etc. because both aminium salts and non-dissociated amines in the aerosol organic phase are detected in their protonated forms during IC measurements. Our usage is consistent with conventions widely adopted in recent studies on atmospheric particulate amines (Liu et al., 2023; Liu et al., 2022; Du et al., 2021).

Table S2: Some water-soluble inorganic ions are "0.0". Please clarify whether these values were below DL or not detected. All chemical components should be reported in consistent units (e.g., in this case, ng m^{-3}); some values are currently given in µg m^{-3}. Indicate whether values represent means or medians and include ranges (min-max). Align the "Component" column to the left. Please also indicate the number of data points available for SYS, NYS, and BS.

Author's response: Thanks for the comment.

We have revised **Table S2** as suggested (see the Appendix).

No value fell below DL or not detected. The "0.0" values were due to very low concentrations that were rounded when expressed in unit of µg m^{-3}.

Line 171: Backward air-mass trajectories should specify the starting hour, given that sampling was conducted over ~24 hours. The authors should also clarify whether the air mass was homogeneous throughout the sampling period (24 hours) and whether it consistently originated from a particular direction. Additional trajectory analyses for individual sampling time points would help address potential air mass variability during the sampling period.

Author's response: Thanks for the comment.

(1) We have added more details in section 2.3 (Lines 187-188). In mobile sampling, each position corresponds to a unique time point.

"**The trajectories were calculated from the position and time point at the beginning of each sampling, with hourly intervals thereafter.**" (Lines 187-188)

(2) We have also revised **Figure S3** and corresponding discussions (Lines 317-319).

"This discrepancy likely reflected secondary formation of amines in aerosols, as well as **the influence of long-range transportation of terrestrial emissions driven by the prevailing East Asia monsoon during spring, particularly to S3 and S12−19 (Figure S3).**" (Lines 317-319)

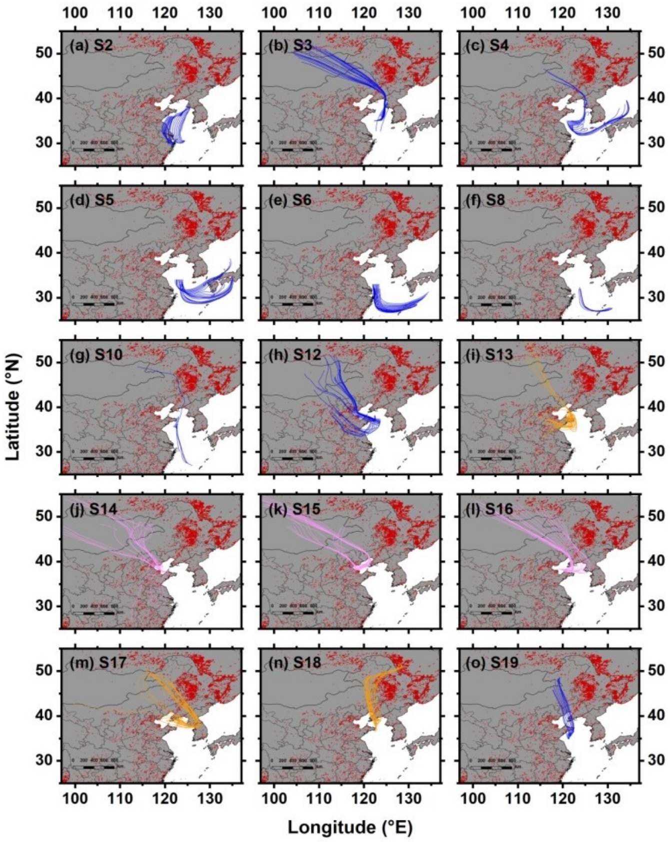

Figure S3. 48 h backward air-mass trajectories starting from the position and time point at the beginning of each sampling, with hourly intervals thereafter. Red dots indicate fire spots during the entire cruise period. Blue, orange, and pink lines represent samples from the SYS, NYS, and BS respectively.

Line 241 & Fig. 2: The ratio seems stable (linear correlation). Is the difference between source areas significant?

Author's response: Thanks for the comment.

We apologize for this error. The differences between source regions were not significant. The discussion of OC/EC ratios has been removed, as it is not essential to the analysis.

Line 249: Amines are strong bases; ammonia is a weak base. I find it difficult to follow why the correlation between amines and ammonium relates to nucleation pathways without having particle number size distribution measurements.

Author's response: Thanks for the comment.

(1) We apologize for the unclear expression. The observed correlation is attributed to the overlapping source profiles of atmosphere NH_3 and gaseous amines, which are precursors of NH_4^+ and particulate amines, respectively. We have revised the paragraph and added relevant information to improve clarity and logic (Lines 257-266).

"Positive correlations were found between NH_4^+ and amines, including MA ($R = 0.78$, $P < 0.01$), DMA ($R = 0.74$, $P < 0.01$), EA ($R = 0.57$, $P < 0.05$), PA ($R = 0.58$, $P < 0.05$), and TMDEA ($R = 0.52$, $P < 0.05$). Aerosol NH_4^+ is formed via the heterogeneous uptake of NH_3, the most abundant alkaline gas in the atmosphere, by acidic aerosols, and exists as ammonium sulfate [$(NH_4)_2SO_4$], ammonium bisulfate (NH_4HSO_4), ammonium nitrate (NH_4NO_3), and ammonium chloride (NH_4Cl) (Behera et al., 2013). **Atmosphere NH_3 shares overlapping source profiles with gaseous amines, including animal husbandry, biomass burning, vehicle emissions, industrial activities, soil, and the ocean. This was inferred as the reason for observed correlations between NH_4^+ and amines in aerosols.**" (Lines 257-266)

(2) The discussion of nucleation has been removed as it is not very relevant to our analysis.

Line 269-270: OP-LW, OP-WP, WP-SP, SNA? Explain please.

Author's response: Thanks for the comment.

(1) OP-LW refers to a mostly organic phase containing little water; OP-WP denotes an organic shell engulfed with a water phase core; WP represents water phase; and SP

indicates solid phase. The hypothesis suggests that amines/NH_4^+ follows the trend OP-LW >> OP-WP > WP or SP, considering both aminium salts and un-protonated amines in atmospheric particles. However, this discussion has been removed, as it relates to new particle formation mechanisms and is not essential to the main results of this study.

(2) SNA is the general term for sulfates, nitrates and ammonium salts. We have removed it and used specific species (Lines 283-285).

"The composition of **NH_4^+, NO_3^-, and SO_4^{2-}** may influence aerosol amines, as they can act as competitors for neutralization and as major reactants in aerosol formation." (Lines 283-285)

Fig 1: Red-orange and blue-lighter blue colors too close to one another. Difficult to distinguish which is which. Could you use another color scheme?

Author's response: Thanks for the comment.

We have revised the color scheme of **Figure 1**.

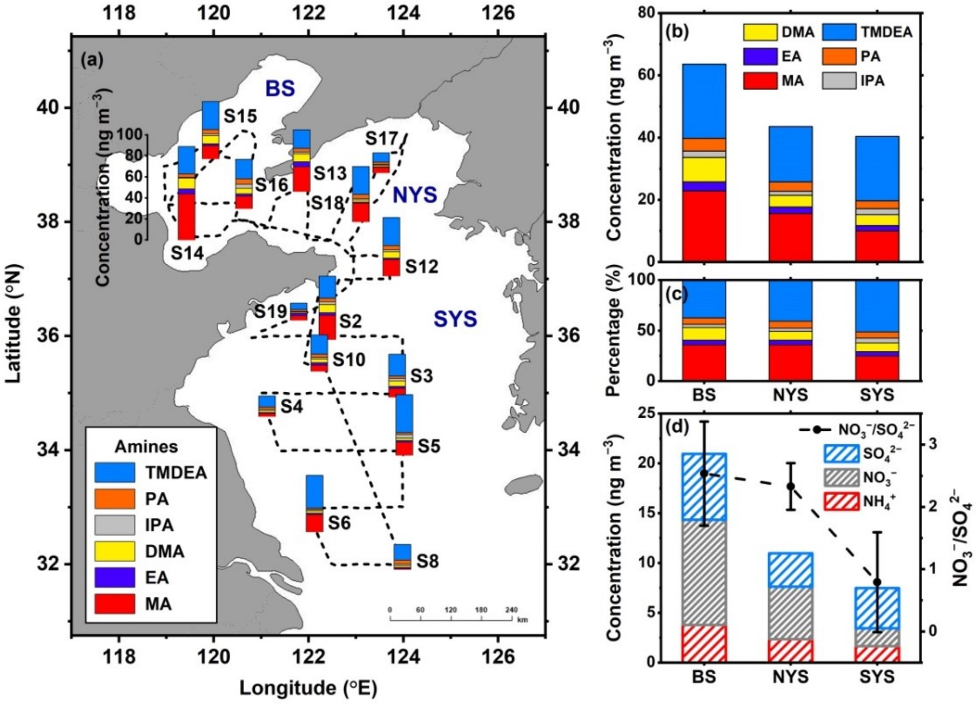

Figure 1. Concentrations of amines in 15 TSP samples (a) collected along the cruise track (black dotted line); average concentrations (b) and relative contributions (c) of amines; and concentrations of NH_4^+, NO_3^-, and SO_4^{2-}, along with NO_3^-/SO_4^{2-} molar ratios (d), in TSP over the SYS, NYS, and BS.

Fig 2 & S5: Define line at 1.0? What is it for? What does the marker size represent?

Author's response: Thanks for the comment.

(1) It is generally accepted that a molar ratio of $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-}) \geq 1$ indicates NH_4^+ fully neutralizes the acidic ions (Cl^-, NO_3^-, and SO_4^{2-}), and a ratio < 1 means a deficiency of NH_4^+. We have removed the line from the Figures to avoid misunderstandings, as these information is already described in the main text. (Lines 285-287).

"The $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$ molar ratios is commonly used to assess whether NH_4^+ fully neutralizes acidic species (Cl^-, NO_3^-, and SO_4^{2-}) in aerosols." (Lines 285-287)

(2) We have added the annotations for symbol size in **Figure 2**.

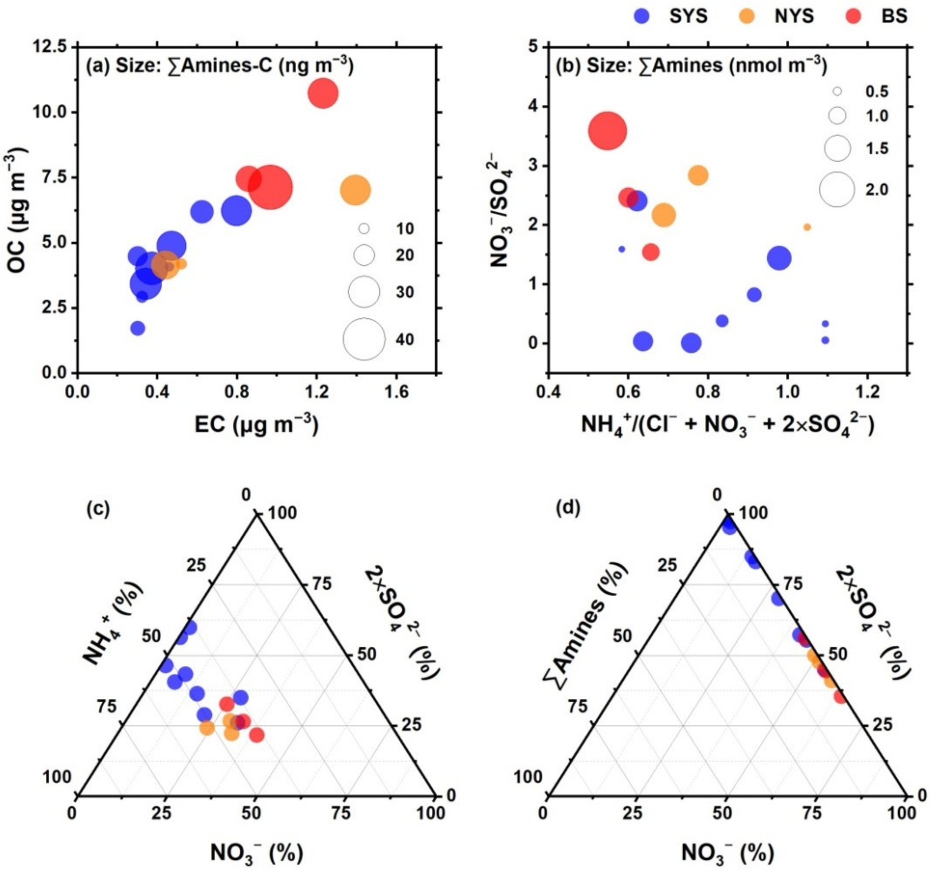

Figure 2. Variations of ∑amines-C with OC and EC concentrations (a); variations of ∑amines molar concentrations with the NO_3^-/SO_4^{2-} and $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$ molar ratios (b); ternary diagram of the molar ratio of NH_4^+, NO_3^-, and SO_4^{2-} (c); and ternary diagram of the molar ratio of ∑amines, NO_3^-, and SO_4^{2-} (d) in TSP over the SYS, NYS, and BS.

Fig S4: Use a colorbar instead of symbol size to represent spatial distributions of amine, and different symbols for location (SYS, NYS and BS).

Author's response: Thanks for the comment.

We have revised **Figure S4** as suggested.

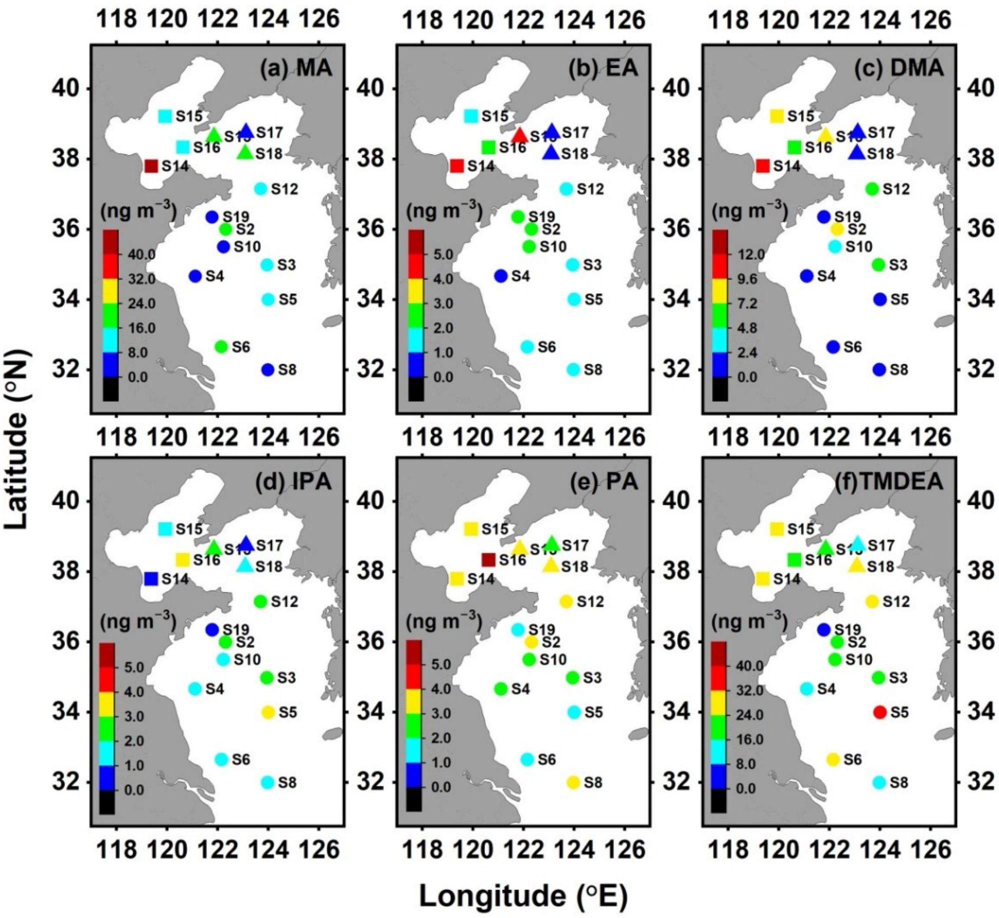

Figure S4. Spatial distributions of MA (a), EA (b), DMA (c), IPA (d), PA (e), and TMDEA (f) in TSP samples over the YS−BS. Circles, triangles, and squares represent samples from the SYS, NYS, and BS, respectively.

Line 330: Replace "6 isoprene SOA" with "Six isoprene SOA"

Author's response: Thanks for the comment.

We have revised as suggested (Line 344).

"In this study, **six isoprene SOA** (SOA_I) tracers, three monoterpene SOA (SOA_M) tracers, and one β-caryophyllene SOA (SOA_C) tracer were measured in TSP over the YS−BS." (Lines 344-346)

Line 402: Replace “Formula” with “Equation” throughout

Author’s response: Thanks for the comment.

We have revised as suggested throughout.

Reviewer #2

This manuscript presented ship-based measurements of low molecular weight amines in aerosol samples from the Bohai Sea and Yellow Sea, China. They discussed the sources and secondary formation pathways of the studied amines based on an extensive chemical dataset, which has not been reported before. They also found that not only primary emissions but secondary formation was important for marine amines, and this was a very important finding. The topic was relevant to atmospheric chemistry and marine–continental interactions, and the manuscript was generally well organized and clearly written. I therefore recommend possible publication after minor revision.

From a presentation perspective, the in-depth discussion occasionally shifts from summarizing observational results to reviewing general chemical mechanisms reported in previous studies, without clearly indicating the added value of the present study. For example, several paragraphs focus on describing nitrate- and sulfate-related reaction pathways, while the specific constraints provided by the this cruise observations are less explicitly highlighted (but this part was very important). Clarifying which statements are directly supported by the measurements and which serve as contextual background would improve readability and focus.

I also suggest to remove all parts related to new particle formation as this part is not relevant for the current study.

Sincerely thanks for the reviewer's comment! We have read carefully, revised and responded to each point. **The modified contents are shown in bold font.**

Author's response:

(1) In the revised manuscript, we have carefully reorganized Section 3.3.3 and Section 3.3.4 to better distinguish between observational results, inferences, and literature-based interpretation. Statements directly supported by our measurements have been clarified, and background information have been streamlined.

(2) Considering all reviewers' comments, we have removed all parts related to new particle formation in results and discussion.

Details:

1. Lines 24–25 "corresponding to the declined influence of terrestrial air masses" Replace "declined" with "decreasing".

Author's response: Thanks for the comment.

We have revised as suggested (Line 25).

"Concentrations of total amines exhibited a north-to-south decrease from the Bohai Sea to the South Yellow Sea, corresponding to the **decreasing** influence of terrestrial air masses." (Lines 23-25)

2. Lines 31–32 "interacting with BSOA formation" Consider rephrasing to clarify whether this refers to concurrent variability or shared environmental/chemical conditions.

Author's response: Thanks for the comment.

We have removed "interacting with BSOA formation" for conciseness in Abstract. The potential interactions among amines, BSOA, and NO_3^- formation processes were discussed in detail in Section 3.3.3, and further clarified in the Conclusions (Line 562)

"MA, EA, and DMA were mainly derived from terrestrial biogenic and non-combustion anthropogenic sources, followed by fossil fuel combustion, with over 50% formed via nitrate-associated secondary formation pathways, **interacting with BSOA formation in the NO_x-involved oxidation of BVOCs**." (Lines 559-562)

3. Lines 44–48 The sentence describing the ubiquity of amines in both the gas and particle phases is relatively long and could be split to improve readability.

Author's response: Thanks for the comment.

We have revised as suggested (Lines 44-48).

"**Low molecular weight amines, such as methylamine (MA), dimethylamine (DMA), trimethylamine (TMA), ethylamine (EA), diethylamine (DEA), and propanamine (PA), are the most common and abundant atmospheric amines. They are ubiquitous in both the gas and particle phases due to high water solubility and strong alkalinity.**" (Lines 44-48)

4. Lines 75–76 "associated with biological activities" Please specify whether this refers to marine biological activity, microbial processes, or biological activity in general.

Author's response: Thanks for the comment.

We have specified to “marine biological activities” (Line 83).

“Elevated concentrations of DMA and TMA are associated with **marine biological activities** (Carpenter et al., 2012; Welsh, 2000) and algal blooms (Müller et al., 2009; Facchini et al., 2008).” (Lines 82-84)

5. Lines 233–234 “suggest that MA, EA, and DMA shared similar sources” Revise verb tense to present tense (“share similar sources”).

Author’s response: Thanks for the comment.

We have revised the manuscript. The observational results and inferences were presented in the past tense consistently (Lines 244-247).

“These results **suggested** that MA, EA, and DMA **might share similar sources** and secondary formation pathways, whereas IPA, PA, and TMDEA were likely influenced by different sources or atmospheric formation processes.”

6. Lines 239–240 Consider revising “generally constitute a small fraction” to “constitute only a minor fraction”

Author’s response: Thanks for the comment.

We have revised as suggested (Lines 250-251).

“Amines, as a subset of water-soluble organic carbon, generally **constitute only a minor fraction** of OC.” (Lines 250-251)

7. Lines 270–272 not clear.

Author’s response: Thanks for the comment.

We have improved the clarity and logic (Lines 283-287).

“**The composition of NH_4^+, NO_3^-, and SO_4^{2-} may influence aerosol amines, as they can act as competitors for neutralization and as major reactants in aerosol formation. The $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$ molar ratios is commonly used to assess whether NH_4^+ fully neutralizes acidic species (Cl^-, NO_3^-, and SO_4^{2-}) in aerosols.**” (Lines 283-287)

8. Lines 284–285 “distinct emission sources and formation mechanisms” Please clarify what “distinct” refers to (e.g., primary vs secondary, marine vs terrestrial, nitrate vs sulfate).

Author’s response: Thanks for the comment.

We have revised as suggested (Lines 298-301).

“Nevertheless, individual amines responded differently to variations in NH_4^+ deficiency and NO_3^-/SO_4^{2-} molar ratios, likely reflecting differences in their **primary sources (terrestrial vs. marine) and formation pathways (nitrate vs. sulfate associated)**.” (Lines 298-301)

9. Lines 550–552 “Different from other amines …” Suggested revision: “In contrast to other amines, TMDEA was predominantly (>60%) generated …”

Author’s response: Thanks for the comment.

We have revised as suggested (Line 565).

“**In contrast to other amines**, TMDEA was mostly (~60%) generated via sulfate-associated secondary formation pathways, and also contributed by primary marine aerosols from sea spray and bubble bursting.” (Lines 565-567)

10. The manuscript would benefit from improvements in language clarity and figure presentation in general.

Author’s response: Thanks for the comment.

We have improve the language clarity and figure presentation in our manuscript.

Appendix

Table S1. Summary of sampling information during the cruise.

Sample ID	Sampling period		Sampling duration (min)	Midpoint position of the sampling period		Sea area	Average ambient temperature (°C)	Average relative humidity (%)	Average wind speed (m s^{-1})
	Start time	End time		Longitude (°E)	Latitude (°N)				
S2	16:30, 28 Mar.	16:30, 29 Mar.	1020	122.32	36.00	SYS	8.2	99.3	6.4
S3	16:38, 29 Mar.	16:20, 30 Mar.	810	123.95	34.98	SYS	9.1	86.3	5.2
S4	17:20, 30 Mar.	15:10, 31 Mar.	1170	121.12	34.67	SYS	8.8	99.2	4.9
S5	15:40, 31 Mar.	15:13, 1 Apr.	1110	124.00	34.00	SYS	12.2	100.1	6.9
S6	15:15, 1 Apr.	15:20, 2 Apr.	1050	122.15	32.65	SYS	12.1	100.3	7.2
S8	15:30, 2 Apr.	20:40, 2 Apr.	450	123.98	32.00	SYS	13.0	99.7	6.6
S10	15:30, 3 Apr.	19:34, 3 Apr.	450	122.23	35.50	SYS	7.8	100.3	11.1
S12	16:30, 7 Apr.	17:41, 8 Apr.	840	123.70	37.15	SYS	6.2	68.9	6.7
S13	19:11, 8 Apr.	18:20, 9 Apr.	1050	121.85	38.63	NYS	7.1	83.4	6.7
S14	09:00, 10 Apr.	07:50, 11 Apr.	810	119.38	37.80	BS	10.9	67.4	7.7
S15	16:53, 11 Apr.	16:53, 12 Apr.	630	119.93	39.22	BS	7.8	67.4	7.5
S16	18:03, 12 Apr.	18:00, 13 Apr.	540	120.63	38.33	BS	8.3	66.6	6.6
S17	18:05, 13 Apr.	20:00, 14 Apr.	1410	123.12	38.75	NYS	6.1	93.2	5.0
S18	14:31, 15 Apr.	14:33, 16 Apr.	810	123.08	38.15	NYS	7.1	79.8	4.7
S19	15:00, 16 Apr.	23:58, 16 Apr.	540	121.78	36.35	SYS	9.1	81.5	5.2

Table S2. Statistical summary of measured chemical components.

Component (ng m^{-3})	SYS (N = 9)	NYS (N = 3)	BS (N = 3)
Amines	40.4 ± 16.4	43.5 ± 17.5	63.6 ± 18.3
MA	10.0 ± 7.0	15.7 ± 7.7	22.8 ± 15.0
EA	1.7 ± 0.6	2.0 ± 1.8	3.0 ± 1.3
DMA	3.5 ± 2.1	3.8 ± 2.6	7.9 ± 2.1
IPA	1.9 ± 0.9	1.3 ± 0.7	2.1 ± 1.4
PA	2.5 ± 0.9	3.0 ± 0.7	4.1 ± 0.7
TMDEA	20.7 ± 9.1	17.8 ± 7.3	23.8 ± 3.7
Water soluble inorganic ions (WSIIs)	8692.1 ± 4363.1	13043.1 ± 6299.9	26912.3 ± 4926.0
Na^+	510.4 ± 455.1	714.7 ± 459.3	1626.9 ± 323.8
NH_4^+	1642.1 ± 657.5	2357.2 ± 989.6	3778.2 ± 481.2
K^+	162.8 ± 70.5	183.3 ± 74.7	481.4 ± 110.4
Mg^{2+}	41.3 ± 38.9	82.9 ± 51.3	271.0 ± 49.4
Ca^{2+}	253.5 ± 199.1	456.6 ± 199.6	1949.9 ± 441.7
F^-	12.3 ± 10.1	28.6 ± 31.2	36.1 ± 7.3
Cl^-	237.6 ± 411.3	602.2 ± 425.1	1582.1 ± 570.7
NO_2^-	4.4 ± 3.5	2.8 ± 2.2	4.3 ± 5.3
NO_3^-	1805.9 ± 2097.6	5255.1 ± 2655.6	10569.0 ± 3189.7
SO_4^{2-}	4021.8 ± 2178.3	3359.6 ± 1544.8	6613.4 ± 893.5
Low molecular organic acids	249.3 ± 124.3	204.0 ± 91.0	247.7 ± 17.0
CHO_2^-	31.8 ± 8.9	38.8 ± 26.2	50.6 ± 10.6
$C_2H_3O_2^-$	29.5 ± 13.5	27.4 ± 10.8	52.1 ± 20.1
$C_4H_4O_4^{2-}$	44.9 ± 36.0	40.5 ± 28.1	58.2 ± 8.3
$C_5H_6O_4^{2-}$	43.7 ± 26.3	47.9 ± 27.4	47.1 ± 2.2
$CH_3O_3S^-$ (MSA^-)	99.5 ± 76.1	49.5 ± 39.9	39.7 ± 8.2
Carbonaceous components (TC)	4670.7 ± 1511.5	5911.3 ± 1775.7	9467.5 ± 1774.8
Organic carbon (OC)	4227.8 ± 1375.7	5126.6 ± 1344.7	8448.9 ± 1627.7
Elemental carbon (EC)	442.9 ± 159.5	784.7 ±431.6	1018.6 ± 156.8
Organic compositions	126.2 ± 74.3	230.6 ± 159.8	474.8 ± 174.6
n-Alkanes (ALK, C_{14}–C_{35})	41.4 ± 22.5	50.6 ± 20.8	93.8 ± 29.1
ALK_{LMW} (C_{20}–C_{26})	22.9 ± 13.8	25.4 ± 10.3	40.3 ± 10.6
ALK_{HMW} (C_{27}, C_{29}, C_{31}, C_{33})	11.2 ± 6.0	16.9 ± 7.4	35.9 ± 13.5
Fatty acids (FA, $C_{14:0}$–$C_{30:0}$)	33.4 ± 15.4	56.4 ± 49.9	67.9 ± 25.3
FA_{LMW} ($\leq C_{19:0}$)	24.1 ± 6.5	29.8 ± 19.6	54.0 ± 20.1
FA_{HMW} ($> C_{19:0}$)	3.6 ± 3.0	10.3 ± 8.3	11.5 ± 4.1
Fatty alcohols (ALC, C_{15}–C_{30})	2.9 ± 1.3	5.0 ± 3.0	8.7 ± 3.2
ALC_{LMW} ($\leq C_{19alc}$)	1.4 ± 0.5	1.0 ± 0.6	1.5 ± 0.4

Component (ng m^{-3})	SYS (N = 9)	NYS (N = 3)	BS (N = 3)
ALC_{HMW} (> C_{19alc})	1.5 ± 0.9	3.9 ± 2.4	7.2 ± 3.0
Polycyclic aromatic hydrocarbons (PAHs)	0.9 ± 0.8	2.0 ± 1.1	3.9 ± 1.5
Hopanes	3.0 ± 3.7	2.4 ± 1.3	4.6 ± 2.5
Steranes	0.5 ± 0.5	0.4 ± 0.2	0.9 ± 0.5
Anhydrosugars	20.0 ± 27.4	46.2 ± 36.7	69.8 ± 22.9
Lignin products	2.0 ± 1.5	4.3 ± 2.4	6.6 ± 1.4
Primary sugars and sugar alcohols	11.8 ± 10.7	37.4 ± 32.0	190.0 ± 97.4
Hydroxy-/polyacids	3.4 ± 2.9	11.5 ± 9.1	7.8 ± 1.7
Aromatic acids	3.4 ± 2.3	7.3 ± 4.1	10.0 ± 2.4
Isoprene SOA (SOA_I)	0.4 ± 0.2	0.9 ± 0.6	1.7 ± 0.8
Monoterpene SOA (SOA_M)	2.1 ± 1.2	4.8 ± 2.8	7.6 ± 3.3
β-Caryophyllene SOA (SOA_C)	0.3 ± 0.3	0.5 ± 0.3	0.8 ± 0.3
2,3-dihydroxy-4-oxopentanoic acid (DHOPA)	0.5 ± 0.3	0.8 ± 0.4	0.6 ± 0.1

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