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Secondary formation dominated low molecular weight amines origins in aerosols over the marginal seas of China

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Abstract. Atmospheric low molecular weight amines play important roles in new particle formation, aerosol properties, and climate. However, the compositions, sources, and secondary formation mechanisms of amines in offshore aerosols remain unclear. Here, an integrated observation of methylamine (MA), ethylamine (EA), dimethylamine (DMA), iso-propanamine (IPA), propanamine (PA), "trimethylamine + diethylamine" (TMDEA), and > 100 other chemical components in total suspended particles was conducted during a spring 2018 research cruise across the Yellow Sea and Bohai Sea, China. Concentrations of total amines exhibited a north-to-south decrease from the Bohai Sea to the South Yellow Sea, corresponding to the declined influence of terrestrial air masses. Source analyses were performed by evaluating the linear relationships between individual amines and specific organic molecular tracers representing primary biogenic sources, higher plant waxes, marine/microbial sources, biogenic secondary organic aerosols (BSOA), biomass burning, and fossil fuel combustion. MA, EA, and DMA were largely influenced by terrestrial biogenic and anthropogenic sources, with the majority (74.0%, 52.6%, and 65.7%, respectively) formed through nitrate-associated secondary formation pathways, interacting with BSOA formation. PA was mainly derived from combustion-related sources, along with terrestrial and marine biogenic contributions. In contrast, the predominate amine, TMDEA, was mostly generated through sulfate-associated secondary formation pathways (61.8%) and contributed by marine emissions, leading to the north-to-south increase in its relative contribution to aerosol amines. The findings highlight distinct potential sources and formation mechanisms for different amines in offshore aerosols, and underscore the importance of multiphase chemistry of amines under varying ambient conditions.

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1 Introduction

Amines, derivatives of ammonia (NH_3) with one or more hydrogen atoms replaced by alkyl or aryl groups, represent an important class of nitrogen-containing organic compounds (Shen et al., 2023; Zhu et al., 2022; Liu et al., 2023). 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, which are ubiquitous in both the gas and particle phases due to high water solubility and strong alkalinity (Ge et al., 2011a, b). These amines are primarily emitted in the gas phase, and mainly occur as protonated cations (aminium salts) in aerosols.

Gaseous amines can be oxidized by atmospheric oxidants (including OH, O_3, and NO_x) (Tang et al., 2013; Nielsen et al., 2012; Murphy et al., 2007), and undergo gas-to-particle conversion through direct dissolution (Liu et al., 2018), acid-base reactions (Liu et al., 2023; Barsanti and Pankow, 2006), and heterogeneous reactions (Pankow, 2015; Chan and Chan, 2013; Qiu and Zhang, 2013), leading to the formation of secondary organic aerosols (SOA) that aggravate air quality and visibility. Gaseous amines and their oxidization products, such as nitrosamines, pose significant risks to human health (Li et al., 2019a; Lee and Wexler, 2013). The multiphase chemistry of atmospheric amines participates in and accelerates new particle formation (Liu et al., 2022; Huang et al., 2022; Yao et al., 2018; Shen et al., 2019), enhances aerosol hygroscopicity (Chu et al., 2015; Gomez-Hernandez et al., 2016), and promotes the activation of cloud condensation nuclei (Tang et al., 2014; Corral et al., 2022; Gomez-Hernandez et al., 2016). Additionally, amines can promote the formation of brown carbon (Marrero-Ortiz et al., 2018; Lin et al., 2015), thereby affecting atmospheric radiation and climate.

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Atmospheric amines originate from diverse natural sources (e.g. ocean, soil, and vegetation) and anthropogenic sources (e.g. animal husbandry, biomass burning, coal combustion, vehicle emissions, composting, waste incineration, industrial activities, and sewage) (Shen et al., 2017; Hemmilä et al., 2018; Feng et al., 2022). The ocean is an important natural source of low molecular weight amines, with emissions mainly driven by biological processes (Calderón et al., 2007; Wang and Lee, 1994). Global modeling (Myriokefalitakis et al., 2010) suggested that amines contribute ~20% of marine SOA, ranking second to dimethylsulfide (DMS). Amines exhibit varying concentrations across different oceans in both seawater and the atmosphere (Violaki and Mihalopoulos, 2010; Gibb et al., 1999; Van Neste et al., 1987). Elevated concentrations of DMA and TMA are associated with biological activities (Carpenter et al., 2012; Welsh, 2000) and algal blooms (Müller et al., 2009; Facchini et al., 2008b). Marine organisms act as both sources and sinks of amines, and the source/sink capability of the ocean varies with ambient conditions (Pinxteren et al., 2019). For instance, TMA can be released from living tissues or during biodegradation and decay, and can also be utilized by microorganisms for energy metabolism (Sun et al., 2019; Köllner et al., 2017; Lidbury et al., 2015). TMA can be biologically oxidized to trimethylamine oxide (TMAO), an osmotic regulatory compound in marine organisms and a precursor of DMA and MA (Chen et al., 2011; Lidbury et al., 2017). The calculated sea-to-air fluxes of DMA at Cape Verde were both positive and negative, whereas those of MA were mostly positive (Pinxteren et al., 2019). Amines in marine aerosols can originate from sea spray (Bates et al., 2012; Gorzelska and Galloway, 1990), bubble bursting (Milne and Zika, 1993), and gas-to-particle conversion through secondary formation (Rinaldi et al., 2010; Facchini et al., 2008b; Facchini et al., 2008a). Most low molecular weight amines in marine

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aerosols were considered to be secondarily formed (Gaston et al., 2013; Dall´osto et al., 2019). For instance, 11−25% of MA, DMA and TMA in the Antarctic sympagic environment originated from primary marine aerosols, whereas 75−89% were incorporated into aerosols after air-sea exchange (Dall´osto et al., 2019). Amines in marine aerosols may also be influenced by inland sources and long-range atmospheric transport (Nielsen et al., 2012). TMA detected in aerosols off the coast of California was associated with inland animal husbandry activities rather than local marine biogenic emissions (Gaston et al., 2013).

Atmospheric low molecular weight amines have been widely reported in urban (Cheng et al., 2020; Chen et al., 2019; Liu et al., 2017), rural (Cheng et al., 2018; Lin et al., 2017), and coastal areas (Liu et al., 2022; Hu et al., 2015; Zhou et al., 2019), but relatively few studies have focused on marine regions of China (Zhou et al., 2019; Yu et al., 2016; Hu et al., 2015). The Bohai Sea (BS) and Yellow Sea (YS) 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). The BS is the northernmost marginal sea of China, surrounded by land on three sides and bordered to the east by the YS. The YS is divided into North Yellow Sea (NYS) and South Yellow Sea (SYS), both semi-open sea areas of the NWPO. Aerosols over the YS−BS are significantly influenced by the transport of terrestrial emissions from northern and eastern China during the prevailing spring East Asia monsoon (Fang et al., 2016). Previous studies on amines in marine aerosols 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). 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). The compositions, sources, and secondary

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formation pathways of amines in offshore aerosols of China remain unclear due to the combined influence of complex terrestrial and marine emissions. Here, spatial variations of amines in aerosols over the YS−BS were investigated through integrated observations of 6 major amines and > 100 other chemical components during a research cruise conducted in spring 2018. The potential sources and secondary formation pathways of different amines were further discussed. The results will enhance understanding of amines in marine aerosols, provide insights into the impact of terrestrial emissions on the atmosphere over the NWPO, and support further research on the gas-to-particle conversion of amines under varying ambient conditions.

2 Methods

2.1 Aerosol sampling

During a Chinese oceanographic cruise over the YS−BS (28 March−16 April 2018), total suspended particles (TSP) were collected on prebaked (450 °C for 6 h) quartz fiber filters using a high-volume air sampler (ASM-1000, Guangzhou; flow rate: 1 m^3 min^{-1}) aboard the *Dong Fang Hong 2* (Figure S1). The sampler was installed windward on the upper deck at the ship bow (~10 m above the sea surface). To avoid contamination from the ship exhaust, sampling was performed only while the vessel was underway. During the sampling period, a total of 15 samples were collected, and 3 field blank filters were prepared by collecting without airflow. The samples were categorized as SYS, NYS, and BS by sampling positions. Real-time navigation and meteorological data, including position, ambient temperature (T), relative humidity (RH), and wind speed (Table S1), were recorded by the onboard monitoring system.

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141 **2.2 Chemical analysis**

142 Low molecular weight amines can be directly separated and quantified using ion
143 chromatography methods (Feng et al., 2020; Place et al., 2017; VandenBoer et al.,
144 2012; Gibb et al., 1995). 6 major protonated amine species, including methylamine
145 (MA), ethylamine (EA), dimethylamine (DMA), iso-propanamine (IPA), propanamine
146 (PA), and the combined species "trimethylamine (TMA) + diethylamine (DEA)"
147 (TMDEA), were measured by a Thermo Fisher Scientific Dionex ICS-5000+ system,
148 as described in detail elsewhere (Yang et al., 2023). Before analysis, a 0.8 cm^2 portion
149 of each sample or blank filter was ultrasonically extracted 3 times with 10−30 mL of
150 ultrapure water for 15 min in an ice-water bath, followed by filtration through a 0.22
151 μm Teflon filter. The analytical precision was better than 10%, and recoveries for all
152 amines ranged from 90% to110%. The method detection limits (MDLs) for MA, EA,
153 DMA, IPA, PA, and TMDEA were 0.4 ng m^{-3}, 0.4 ng m^{-3}, 0.5 ng m^{-3}, 0.7 ng m^{-3}, 1.1
154 ng m^{-3}, and 2.9 ng m^{-3}, respectively.

155 To provide a comprehensive characterization, other key chemical components were
156 also analyzed, including water-soluble inorganic ions (WSIIs; Na^+, NH_4^+, K^+, Mg^{2+},
157 Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}, etc.), low molecular weight organic acids (CHO_2^-, $C_2H_3O_2^-$,
158 $C_4H_4O_4^{2-}$, $C_5H_6O_4^{2-}$, $CH_3O_3S^-$/MSA^-, etc.), carbonaceous components (TC, OC, and
159 EC), and organic compositions (polar and nonpolar). Detailed methodologies had been
160 described elsewhere (Fan et al., 2019; Cao et al., 2024), and the measurement results
161 were summarized in Table S2.

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2.3 Auxiliary data

Average chlorophyll *a* (Chl *a*) concentrations in seawater during the sampling period were retrieved from combined Aqua-MODIS and Terra-MODIS datasets (https://oceancolor.gsfc.nasa.gov/) using ArcGIS software (Figure S2). Fire spot information was obtained from the Fire Information for Resource Management System (FIRMS, https://firms.modaps.eosdis.nasa.gov/). Based on the archived Global Data Assimilation System (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/) meteorological data, 48 h backward air-mass trajectories at 200 m above ground level were calculated using the Hybrid Single-particle Lagrangian Integrated Trajectory (HYSPLIT) model, and subsequently processed with TrajStat software (Figure S3).

3 Results and discussion

3.1 Overview of amines in marine aerosols

During the research cruise over the YS−BS from 28 March to 16 April 2018, total concentrations of MA, EA, DMA, IPA, PA, and TMDEA (∑amines) in TSP ranged from 16.2 ng m^{-3} to 89.1 ng m^{-3} (Figure 1). Lower ∑amines concentrations were observed over the SYS and NYS, averaging 40.4 ± 16.4 ng m^{-3} and 43.5 ± 17.5 ng m^{-3}, respectively, and higher concentrations occurred over the BS, averaging 63.6 ± 18.3 ng m^{-3}. Concentrations of other chemical components, including total WSIIs, TC, and total measured organic compositions, exhibited a similar spatial pattern (SYS < NYS < BS; Table S2).

TMDEA was the predominant amine species in TSP over the YS−BS, with concentrations ranging from 6.1 ng m^{-3} to 36.3 ng m^{-3} (Figure S4) and averages of 20.7 ± 9.1 ng m^{-3}, 17.8 ± 7.3 ng m^{-3} and 23.8 ± 3.7 ng m^{-3} over the SYS, NYS and

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BS, respectively. TMDEA accounted for a larger fraction of ∑amines over the SYS (51.2%) than the NYS (40.8%) and BS (37.4%). Compared with previous studies (Table. S3), the average TMDEA concentration in TSP over the YS−BS during March−April 2018 (20.7 ± 8.2 ng m^{-3}) was comparable to values reported for $PM_{0.056–10}$ during June−July 2016 (21.0 ng m^{-3}) and August 2015 (18.6 ng m^{-3}) (Xie et al., 2018), for $PM_{2.5}$ during January–February 2018 at coastal Qingdao (a port city surrounded by the YS and BS, 19.3 ng m^{-3}) (Liu et al., 2022), and for PM_1 in July 2007 off the central coast of California, USA (22.0 ng m^{-3}) (Sorooshian et al., 2009). These values were higher than those measured in $PM_{0.056–10}$ at coastal Qingdao (9.0 ng m^{-3}) (Xie et al., 2018) and in $PM_{2.5}$ at Huaniao Island (an adjacent island of China, 8.7 ng m^{-3}) (Zhou et al., 2019) during August 2016, and markedly higher than those in aerosols from other marine regions, including the Arabian Sea (Gibb et al., 1999), the East Mediterranean (Violaki and Mihalopoulos, 2010), the North Atlantic (Facchini et al., 2008a), and the tropical Atlantic (Pinxteren et al., 2019; Müller et al., 2009). In aerosols over the SYS and NYS−BS, TMDEA concentrations in TSP during spring (20.7 ng m^{-3} and 20.8 ng m^{-3} in March−April 2018) were lower than those measured in PM_{10} during summer (45.6 ng m^{-3} in August 2015 and 28.8 ng m^{-3} in August−September 2015) (Yu et al., 2016).

MA, the second most abundant amine species (range: 0.9−44.0 ng m^{-3}), exhibited average concentrations of 22.8 ± 15.0 ng m^{-3} and 15.7 ± 7.7 ng m^{-3} in TSP over the BS and NYS, contributing 35.9% to ∑amines. Relatively lower MA concentrations (10.0 ± 7.0 ng m^{-3}) and a smaller proportion of MA to ∑amines (24.9%) were observed over the SYS compared with the NYS−BS. The spatial pattern of MA contributions to ∑amines was opposite to that of TMDEA. The average MA concentration in TSP over the YS−BS (13.7 ± 10.5 ng m^{-3}) was higher than those

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observed in $PM_{2.5}$ at coastal Qingdao (8.5 ng m^{-3}, winter 2018) (Liu et al., 2022), in $PM_{0.9}$ over the Arabian Sea (3.2−3.7 ng m^{-3}) (Gibb et al., 1999), and in PM_1 over the tropical Atlantic (0.2 ng m^{-3}) (Pinxteren et al., 2019), but comparable to that in $PM_{2.5}$ at Jeju Island, South Korea (13.5 ng m^{-3}) (Yang et al., 2004).

DMA concentrations ranged from 1.3 ng m^{-3} to 10.4 ng m^{-3}, with averages of 3.5 ± 2.1 ng m^{-3}, 3.8 ± 2.6 ng m^{-3}, and 7.9 ± 2.1 ng m^{-3} in TSP over the SYS, NYS, and BS, respectively. Higher DMA contributions to ∑amines were found over the BS (12.4%) than the NYS (8.7%) and SYS (8.6%). DMA concentrations in aerosols during spring 2018 over the YS−BS (4.4 ± 2.8 ng m^{-3}) were markedly lower than those observed in winter 2018 at coastal Qingdao (58.7 ng m^{-3}) (Liu et al., 2022), in summer 2015/2016 over the YS−BS (23.9 ng m^{-3} and 50.6 ng m^{-3}) (Xie et al., 2018), and in winter 2013 over the SYS (19.8 ng m^{-3}) (Yu et al., 2016). EA (0.6−4.8 ng m^{-3}), IPA (0.5−3.9 ng m^{-3}), and PA (1.3−5.1 ng m^{-3}) constituted a relatively small fraction of ∑amines (7.3−28.2%), with average concentrations of 2.0 ± 1.2 ng m^{-3}, 1.8 ± 1.0 ng m^{-3}, and 2.9 ± 1.0 ng m^{-3} in TSP over the YS−BS, respectively. EA concentrations in aerosols over the BS (3.0 ± 1.3 ng m^{-3}, March−April 2018) were comparable to those in $PM_{2.5}$ at coastal Qingdao (2.7 ng m^{-3}, January–February 2018) (Liu et al., 2022) and at Jeju Island, South Korea (3.1 ng m^{-3}, March–April 2001) (Yang et al., 2004). Comparable data for EA, IPA and PA concentrations in marine aerosols were currently limited. Strong positive correlations were observed among MA, EA, and DMA ($R = 0.73–0.77$, $P < 0.01$), whereas no statistically significant correlation ($P > 0.05$) exhibited between IPA, PA or TMDEA and other amine species. These results suggest that MA, EA, and DMA shared similar sources and secondary formation pathways, whereas IPA, PA, and TMDEA likely originated from different sources or undergo distinct atmospheric formation processes.

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3.2 Relative contributions of amines in TSP over the YS−BS

Amines, as a subset of water-soluble organic carbon, generally constitute a small fraction of OC. Both OC and EC concentrations in TSP decreased significantly from the BS to the NYS and SYS (Figure 2). Higher OC/EC ratios were observed in samples from the SYS (9.8 ± 2.3), which were more strongly influenced by marine air masses (Figure S3), compared with the BS−NYS (7.9 ± 1.4; Figure S5). The high OC/EC ratios related to reduced contributions from terrestrial combustion sources and enhanced OC emissions from marine microorganisms. The ∑amines-C/OC ratios ranged from 2.1‰ to 8.8‰, with an average of 4.9 ± 2.1‰ in TSP over the YS−BS. These ratios were relatively higher in aerosols over the SYS (5.4 ± 2.2‰) than the NYS (4.4 ± 1.7‰) and BS (4.0 ± 1.4‰).

Low molecular weight amines can act as strong bases even in the presence of NH_3 (Sorooshian et al., 2008). They share similar emission sources and gas-to-particle conversion pathways to some extent, as indicated by the correlations between NH_4^+ and 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 ($R = 0.52$, $P < 0.05$). The molar ratios of amines to NH_4^+ were calculated to evaluate their relative contributions to the neutralization of acidic species in aerosols (Hu et al., 2015). The ∑amines/NH_4^+ molar ratios ranged from 4.8‰ to 17.0‰ (8.7 ± 3.0‰) in TSP over the YS−BS, and were 9.7 ± 3.4‰, 7.6 ± 0.8‰, and 6.8 ± 1.8‰ over the SYS, NYS, and BS, respectively. The spatial pattern of ∑amines/NH_4^+ molar ratios (SYS > NYS > BS) was consistent with that of the ∑amines-C/OC ratios, indicating a north-to-south enhancement in the relative contributions of amines to aerosol composition over the YS−BS.

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The ∑amines/NH_4^+ molar ratios calculated in this study were of the same order of magnitude as those reported previously (Xie et al., 2018; Yu et al., 2016). Overall, amines yield a negligible contribution to the neutralization of acidic species in TSP compared to NH_4^+, which is reasonable given the much higher atmospheric concentrations of NH_3 relative to gaseous amines (Zheng et al., 2015; You et al., 2014; Ge et al., 2011a, b). However, amines may play a more important role in neutralizing acidic species in submicron particles, particularly in the presence of organic compounds, as suggested by the size-dependent amines/NH_4^+ ratio hypothesis (amines/$NH_4^+_{OP\text{-}LW}$ >> amines/$NH_4^+_{OP\text{-}WP}$ > amines/$NH_4^+_{WP\ or\ SP}$) (Xie et al., 2018). The secondary formation of SNA (NH_4^+, NO_3^-, and SO_4^{2-}) interacts with that of amines, thereby influencing their concentrations and compositions in aerosols. The $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$ molar ratios in TSP over the YS−BS were mostly < 1 (0.8 ± 0.2; Figure 2 and Figure S5), indicating NH_4^+ deficiency in aerosols, and the NH_4^+ deficiency was more markedly over the BS (0.6 ± 0.0) than the YS (0.8 ± 0.2). The NO_3^-/SO_4^{2-} molar ratios in TSP over the SYS were 0.8 ± 0.8, significantly lower than those over the NYS (2.3 ± 0.4) and BS (2.5 ± 0.8), suggesting that SO_4^{2-} was the dominate acidic species in SYS aerosols, whereas NO_3^- dominated in NYS and BS aerosols. The characteristics of SNA in NYS aerosols were intermediate between those over the BS and SYS, consistent with the regional variations in concentrations and compositions of amines. Molar concentrations of ∑amines increased with the NH_4^+ deficiency [indicated by $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$ ratios; $R = -0.57$, $P < 0.05$] and with the NO_3^-/SO_4^{2-} ratios ($R = 0.56$, $P < 0.05$), particularly in BS aerosols. Nevertheless, individual amines responded differently to variations in NH_4^+ deficiency and NO_3^-/SO_4^{2-} molar ratios, reflecting their distinct emission sources and formation mechanisms.

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3.3 Source analysis of amines in TSP over the YS−BS

3.3.1 Biogenic sources

On a global scale, the ocean is a major source of gaseous methylamines (fluxes: TMA > MA >> DMA) (Van Neste et al., 1987; Schade and Crutzen, 1995). Intensive ocean farming is widespread in the coastal areas of the YS−BS (Hu et al., 2015), where marine biogenic sources, including fish emission (Namieśnik et al., 2003), biodegradation of nitrogen-containing materials, and decay process (Calderón et al., 2007) may release gaseous amines into the atmosphere. The concentration of Chl *a* in surface seawater is an indicator of phytoplankton biomass and thus reflects the intensity of marine biogenic emissions to some extent. Significantly higher Chl *a* concentrations were observed in the BS than in the YS, with relatively elevated values in near shore areas (Figure S2). The spatial distribution of ∑amines in TSP over the YS−BS was broadly consistent with, though not identical to, the Chl *a* concentrations in surface seawater. This was attributed to secondary formation processes of amines in aerosols, as well as the influence of long-range transport of terrestrial emissions driven by the prevailing East-Asia monsoon during spring, particularly to S10, S12, and S14−18 (Figure S3).

MA, EA, and DMA exhibited positive linear relationships with the total concentrations of primary sugars and sugar alcohols (Figure 3 a−c and Table S4), which mainly originate from primary biogenic sources such as bacteria, pollen, and animal or plant debris (Li et al., 2019b). These primary biogenic sources can be either marine or terrestrial. Fungal spore OC and plant debris OC were estimated from the concentrations of mannitol and arabitol (Bauer et al., 2008), and glucose (Zheng et al.,

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2018), respectively. Significant positive correlations were observed between MA, EA, and DMA and fungal spore OC, plant debris OC, and several individual primary sugars and sugar alcohols, including trehalose, α-fructose, and sucrose ($R > 0.50$, $P < 0.05$). DMA exhibited the strongest correlation with trehalose ($R = 0.71$, $P < 0.01$), which is abundant in microorganisms, algae, plants, and invertebrates, and also acts as an indicator of re-suspended dust (Medeiros et al., 2006; Simoneit et al., 2004). In addition, MA, DMA, and PA were correlated with high molecular weight *n*-alkanes (ALK_{HMW}; C_{27}, C_{29}, C_{31} and C_{33}) and high molecular weight fatty alcohols (ALC_{HMW}; > C_{19alc}; Figure 3 d−e and Table S4). ALK_{HMW} (Rogge et al., 1993), ALC_{HMW} (Simoneit et al., 1991), and high molecular weight fatty acids (FA_{HMW}, > $C_{19:0}$) (Haque et al., 2019) are tracers of higher plant waxes, emitted by terrestrial vegetation. A significant positive relationship was also observed between PA and low molecular weight fatty acids (FA_{LMW}; ≤ $C_{19:0}$; Figure 3 f), which can be emitted by microorganisms, phytoplankton, or bacteria (Haque et al., 2019) and are considered tracers of marine/microbial sources in marine aerosols. These results indicate that amines (MA, EA, DMA, and PA) in TSP over the YS−BS were contributed by biogenic sources. MA and DMA were largely influenced by terrestrial biogenic emissions, whereas PA was affected by both terrestrial and marine biogenic sources.

Atmospheric biogenic secondary organic aerosols (BSOA) are formed through the photochemical oxidation of biogenic volatile organic compounds (BVOCs) with O_3, OH and NO_x (Ng et al., 2011). 6 isoprene SOA (SOA_I) tracers, 3 monoterpene SOA (SOA_M) tracers, and 1 *β*-caryophyllene SOA (SOA_C) tracer were measured in TSP over the YS−BS. Biogenic SOC derived from isoprene, monoterpene, and *β*-caryophyllene was estimated using the tracer-based method (Kang et al., 2018; Kleindienst et al., 2007). Significant positive linearity were observed between MA

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and both isoprene SOC and monoterpene SOC (Figure 3 g–h). Among the 6 SOA_I tracers, MA exhibited stronger correlations with 2-methyltetrols (2-MTLs; $R = 0.74$, $P < 0.01$) and C_5-alkene triols ($R = 0.66$, $P < 0.01$) than with 2-methylglyceric acid (2-MGA; $R = 0.64$, $P < 0.05$). DMA also showed a positive correlation with isoprene SOC ($R = 0.55$, $P < 0.05$), only driven by its association with 2-MTLs ($R = 0.59$, $P < 0.05$). Among the 3 SOA_M tracers, significant correlations were found between pinonic acid and MA ($R = 0.73$, $P < 0.01$), EA ($R = 0.52$, $P < 0.05$), and DMA ($R = 0.58$, $P < 0.05$), as well as between pinic acid with MA ($R = 0.59$, $P < 0.05$). In addition, a positive linearity was observed between only PA and β-caryophyllene SOC ($R = 0.67$, $P < 0.01$; Figure 3 i). These findings supported that MA, EA, DMA and PA shared common emission sources with BSOA precursors and/or interact with BSOA formation processes. High concentrations of amines and biomarkers were simultaneously observed in the BS and NYS aerosols, and amines in SYS aerosols reached moderate levels even under low biomarker concentrations (Figure 3). These indicated that terrestrial biogenic emissions contributed more substantially to amines in aerosols over the BS and NYS than the SYS.

3.3.2 Anthropogenic sources

Anthropogenic sources were another important potential contributor to atmospheric amines. These sources can be categorized into combustion-related sources (e.g., biomass burning, coal combustion, vehicle emissions, and waste incineration) and non-combustion sources (e.g., animal husbandry, composting, industrial activities, sewage, and septic system). The concentrations of EA ($R = 0.61$, $P < 0.05$) and DMA ($R = 0.72$, $P < 0.01$) in TSP over the YS–BS showed synchronous increases with EC,

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indicating the influence of combustion emissions. Levogluctosan (Lev) is a well-established tracer for biomass burning (Li et al., 2019b). The concentrations of Lev derived from biomass burning (Lev_{bb}) were estimated using Lev and non-sea-salt K^+ (nss-K^+ = K^+ − 0.037 × Na^+), considering the atmospheric degradation of Lev and that ~25% of Lev originated from other sources [Lev_{bb} = 0.75 × Lev × nss-K^+/(0.18 × Lev + 0.08 × nss-K^+)]. Biomass burning was not a major source of MA, EA, and DMA in marine aerosols (Figure 3 j), but it contributed substantially to PA, as indicated by the positive linear relationships between PA and both Lev_{bb} and lignin products (Figure 3 k–l). The most significant contributor to PA from biomass burning might be conifer burning (the second-largest portion of total biomass burning), according to the correlations of PA and individual lignin products: 4-hydroxybenzoic acid (4-HBA; a herbs burning marker and the dominate lignin product in this study; R = 0.52, P < 0.05), vanillic acid (VA; a softwood and hardwood burning marker; R = 0.67, P < 0.01), syringic acid (SA; also a softwood and hardwood burning marker; R = 0.60, P < 0.05), and dehydroabietic acid (DA; a conifer burning marker; R = 0.71, P < 0.01). MA (R = 0.57, P < 0.05) and DMA (R = 0.54, P < 0.05) in TSP over the YS–BS exhibited positive correlations with polycyclic aromatic hydrocarbons (PAHs), suggesting potential contributions from fossil fuel combustion (Table S4). PA showed a stronger association with combustion-related sources than other amines, as evidenced by its correlations with multiple fossil fuel combustion tracers (Figure 3 m–o), including low molecular weight n-alkanes (ALK_{LMW}; C_{20}–C_{26}; R = 0.67, P < 0.01), PAHs (R = 0.63, P < 0.05), hopanes (R = 0.55, P < 0.05), and steranes (R = 0.57, P < 0.05).

Emissions of amines (MA, DMA, and TMA) from non-combustion anthropogenic sources, such as composting, sewage, and septic system, are largely linked to

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biodegradation process. Therefore, the contribution of non-combustion anthropogenic sources to amines might be encompassed within the primary biogenic sources category. IPA did not show any correlation with organic molecular tracers in TSP over the YS−BS. Given its widespread use in industrial production (e.g., pesticides, pharmaceuticals, dye intermediates, emulsifiers, detergents, surfactants, and textile additives), IPA might be emitted in particulate form from specific industrial activities (Ge et al., 2011b).

3.3.3 Secondary formation of MA, EA, DMA, and PA

Significant correlations were observed between MA, EA, DMA, and PA with Cl^- and NO_3^- (Figure 4). The intercepts of the regression lines for MA, EA, and DMA against Cl^- or NO_3^- were lower than those obtained with organic molecular tracers of primary sources (Figure 3 and Figure S6), indicating the substantial contribution from secondary formation. Direct dissolution of gaseous amines into aerosol liquid water (Formula 1) is a key step in the gas-to-particle conversion of MA (CH_3NH_2), EA ($CH_3CH_2NH_2$), and DMA [$(CH_3)_2NH$], all of which are highly water-soluble. Low T, high aerosol acidity, and high atmospheric liquid water content favor this process. Gaseous amines may also undergo directly uptake onto acidic particle surfaces (Formula 2) (Yin et al., 2011), which is particularly important under the NH_4^+ deficiency observed in BS aerosols. Acid-base reactions between amines and inorganic acids occur in both gas and aqueous phases. Protonated amines (aminium salts), including MA ($CH_3NH_3^+$), EA ($CH_3CH_2NH_3^+$), DMA [$(CH_3)_2NH_2^+$], and PA ($CH_3CH_2CH_2NH_3^+$), in TSP over the YS−BS could be formed through reactions with atmospheric HCl and HNO_3 (Formula 3−4). Atmospheric CH_3COOH also contributed

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to the formation of MA, EA and DMA in TSP (Figure 4). As NO_3^- concentrations were significantly higher than those of Cl^- and $C_2H_3O_2^-$ in TSP (Table S2), reactions with HNO_3, along with displacement reactions between gaseous amines and NH_4NO_3 (Formula 5) (Bzdek et al., 2010), were the dominate pathway for the secondary formation of MA, EA, DMA and PA in TSP over the YS−BS. The partitioning of amines into aerosols was further promoted by low T, high aerosol acidity, and high RH conditions in the dynamic solid/aqueous/gas equilibrium.

$$R_nNH_{3-n(g)} + H_2O_{(l)} \leftrightarrow R_nNH_{4-n} \cdot OH_{(aq)} \leftrightarrow R_nNH_{4-n(aq)}^+ + OH_{(aq)}^- \quad (1)$$

$$R_nNH_{3-n(g)} + H_3O_{(surface)}^+ \leftrightarrow R_nNH_{4-n(surface)}^+ + H_2O \quad (2)$$

$$R_nNH_{3-n(g)} + HCl_{(g)} \leftrightarrow R_nNH_{4-n}Cl_{(s)} \leftrightarrow R_nNH_{4-n(aq)}^+ + Cl_{(aq)}^- \quad (3)$$

$$R_nNH_{3-n(g)} + HNO_{3(g)} \leftrightarrow R_nNH_{4-n}NO_{3(s)} \leftrightarrow R_nNH_{4-n(aq)}^+ + NO_{3(aq)}^- \quad (4)$$

$$R_nNH_{3-n(g)} + NH_4NO_{3(s)} \rightarrow (R_nNH_{4-n})NO_{3(s)} + NH_{3(g)} \uparrow \quad (5)$$

Nitrate-associated secondary formation contributions were estimated from the average amines concentrations weighted by NO_3^- concentrations and the regression intercepts (Figure S6). These contributions to ∑amines in TSP were highest over the BS (43.0%), followed by the NYS (33.8%) and SYS (21.8%). Among individual amines, nitrate-associated secondary formation contributed most to MA (BS: 79.3%; NYS: 70.5%; SYS: 57.2%), followed by DMA (BS: 73.3%; NYS: 48.3%; SYS: 55.5%), EA (BS: 61.2%; NYS: 47.2%; SYS: 29.0%), and PA (BS: 41.1%; NYS: 30.5%; SYS: 25.2%). PA might also be directly emitted in particulate form or condense into aerosols after emission due to its relatively higher boiling point (47.8°C) compared with MA (−6.3°C), EA (16.6°C), and DMA (7.4°C).

Interactions might exist among the secondary formation of amines (MA, EA, DMA, and PA), BSOA, and nitrate in the atmosphere. NO_x, emitted by soil, biogenic

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activities, and combustion sources, are important precursors for both BSOA and atmospheric HNO_3, which subsequently forms nitrate aerosols. These were supported by the significant positive correlations between NO_3^- and SOA_I ($R = 0.88$, $P < 0.01$), SOA_M ($R = 0.86$, $P < 0.01$), and SOA_C ($R = 0.64$, $P < 0.05$). The formation of MA and DMA in aerosols could occur under low NO_x conditions, as evidenced by their stronger correlations (Text S1) with 2-MTLs or C_5-alkene triols (products of isoprene photochemical oxidation under low NO_x conditions) (Zheng et al., 2018; Zhang et al., 2011) than with 2-MGA (products of isoprene aqueous-phase oxidation under high NO_x conditions) (He et al., 2018). Strong atmospheric photo-oxidation conditions generally accelerates the gas-phase degradation of amines (Lee and Wexler, 2013) due to their reactions with OH, O_3 and NO_x [nitrate radical (NO_3) and HONO (HONO ↔ NO + NO_2 + H_2O)], thereby reducing the formation of particle-phase aminium salts. BVOCs, precursors of BSOA, generated HNO_3 via the "NO_3 + HC" pathway, further enhancing the formation of aminium nitrates. Meanwhile, BSOA formation consumed atmospheric oxidants, which reduced the degradation of gaseous amines to some extent. The presence of an organic phase also increased the competitiveness of amines relative to NH_4^+ in aerosols (Xie et al., 2018). In addition, the gas-to-particle conversion of amines promoted particle nucleation and growth, providing more hygroscopic particulate surfaces that facilitate further BSOA formation.

During the cruise, lower average T and RH were observed over the BS (9.0℃, 67.2%) and NYS (6.7℃, 85.7%) compared to the SYS (9.5℃, 91.8%). RH over the YS−BS generally (90%) remained at a high level (> 61.1%, median: 87.6%). The relatively abundant acidic species and lower T over the BS and NYS favored the partitioning of MA, EA, DMA, and PA into the particle phase, compared with the conditions over the SYS.

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3.3.4 Secondary formation of TMDEA

TMDEA in TSP over the YS−BS showed no correlation with the measured organic molecular tracers, Cl^-, or NO_3^-, but exhibited significant positive linear relationships with SO_4^{2-}, $C_4H_4O_4^{2-}$, and $C_5H_6O_4^{2-}$ (Table S4, Figure 4 and Figure S6). Compared with other amines, a larger fraction of TMDEA originated from marine sources, as suggested by its relatively high concentrations and proportions in TSP over the SYS. Previous studies reported that the global flux of TMA from the ocean was 1.6 times that of MA (Schade and Crutzen, 1995), and DEA has also been detected in seawater and marine aerosols at the tropical Atlantic (Pinxteren et al., 2019). Marine sources were an important contributor of gaseous TMDEA over the YS−BS. In addition, terrestrial vegetation and non-combustion anthropogenic sources (mainly including septic system, animal husbandry, composting, and sewage) also contributed gaseous TMDEA (Zhu et al., 2022; Ge et al., 2011b). However, the concentrations of TMDEA in aerosols were constrained by its gas-to-particle conversion efficiency.

Reactions between TMDEA and NO_3 lead to the formation of non-aminium-salt SOA (Price et al., 2016; Price et al., 2014). Taking TMA [$(CH_3)_3N$] as an example, the reactions (Formula 6–7) produce HNO_3 and peroxy radicals, which subsequently undergo hydrogen rearrangement to generate particle-phase oligomers. This pathway not only yields atmospheric HNO_3 through the "NO_3 + HC" mechanism but also consumes part of the gaseous TMDEA emitted from primary sources. Uptake onto acidic particle surfaces (Formula 2) is 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 was not inclined to present as chloride and nitrate, attributing to

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the much lower competitiveness of TMA in forming chloride and nitrate (reflected by dissociation constants) relative to MA, EA, DMA, and NH_3 (Ge et al., 2011a). Instead, acid-base reactions of TMA and DEA with H_2SO_4 (Formula 8), as well as displacement reactions with $(NH_4)HSO_4$ and $(NH_4)_2SO_4$ (Formula 9−10), were the dominate pathways for the secondary formation of TMDEA in TSP over the YS−BS. These findings were consistent with previous studies that TMA had been proofed to preferentially nucleate with H_2SO_4 (Johnson and Jen, 2023), and irreversible reactive uptake of gaseous ethylamines by H_2SO_4 had been observed, with DEA exhibiting the highest uptake coefficient (Yin et al., 2011). Sulfate-associated secondary formation contributed 61.4%, 55.8%, and 63.4% (estimated by average TMDEA concentrations weighted by SO_4^{2-} concentrations and the regression intercepts, Figure S6) to TMDEA in TSP over the BS, NYS, and SYS, respectively, corresponding to 23.0%, 22.8%, and 32.5% to ∑amines. These contributions increased with average T, which were higher over the SYS, followed by the BS and NYS. Dicarboxylic acids ($C_4H_4O_4$ and $C_5H_6O_4$) also contributed to the secondary formation of TMDEA in TSP over the YS−BS through reactions similar to Formula 8.

$$(CH_3)_3N_{(g)} + \cdot NO_{3(g)} \rightarrow \cdot CH_2N(CH_3)_{2(g)} + HNO_{3(g)} \quad (6)$$

$$\cdot CH_2N(CH_3)_{2(g)} + O_{2(g)} \rightarrow \cdot OOCH_2N(CH_3)_{2(g)} \quad (7)$$

$$R_nNH_{3-n(g)} + H_2SO_{4(g)} \leftrightarrow (R_nNH_{4-n})_2SO_{4(s)} \leftrightarrow R_nNH^+_{4-n(aq)} + SO^{2-}_{4(aq)} \quad (8)$$

$$R_nNH_{3-n(g)} + (NH_4)HSO_{4(s)} \rightarrow (R_nNH_{4-n})(NH_4)SO_{4(s)} \rightarrow (R_nNH_{4-n})_2SO_{4(s)} + NH_{3(g)}\uparrow \quad (9)$$

$$R_nNH_{3-n(g)} + (NH_4)_2SO_{4(s)} \rightarrow (R_nNH_{4-n})(NH_4)SO_{4(s)} + NH_{3(g)}\uparrow \rightarrow (R_nNH_{4-n})_2SO_{4(s)} + NH_{3(g)}\uparrow \quad (10)$$

Significant positive correlations were observed between non-sea-salt sulfate

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(nss-SO_4^{2-} = SO_4^{2-} − 0.2516 × Na^+) and the dicarboxylates $C_4H_4O_4^{2-}$ and $C_5H_6O_4^{2-}$ (R = 0.78 and 0.66, P < 0.01), indicating that these species shared similar potential terrestrial anthropogenic or marine biogenic origins (Miyazaki et al., 2010; Mochida et al., 2003). Molar concentrations of biogenic-SO_4^{2-} were estimated from T (Table S1) and MSA^-, as both MSA^- and SO_4^{2-} are oxidation products of DMS emitted from marine biogenic sources (Nakamura et al., 2005; Bates et al., 1992). Anthropogenic-SO_4^{2-} was determined by subtracting biogenic-SO_4^{2-} from nss-SO_4^{2-}. Biogenic-SO_4^{2-} accounted for 11.1% of total SO_4^{2-} in TSP over the SYS, markedly higher than the contributions over the NYS (4.3%) and BS (2.1%), yet still representing a minor fraction relative to anthropogenic-SO_4^{2-}. TMDEA in TSP over the YS−BS was predominantly taken up by anthropogenic sulfate aerosols.

High concentrations of TMDEA, SO_4^{2-}, $C_4H_4O_4^{2-}$, and $C_5H_6O_4^{2-}$ were simultaneously observed in S5 and S6 over the SYS, along with relatively high marine biogenic contributions (Biogenic-SO_4^{2-}/SO_4^{2-}: 11.2% and 10.3%). NH_4^+ deficiency [NH_4^+/(Cl^- + NO_3^- + 2*SO_4^{2-}): 0.8 and 0.6], high T (12.2°C and 12.1°C), high wind speed (6.9 m s^{-1} and 7.2 m s^{-1}), and saturated water vapor (RH = 100%) conditions were also found in S5 and S6 (Table S1). Under high RH conditions, more amines partition into aqueous aerosols through direct dissolution, favoring the formation of aminium salts. However, high T can shift the solid/aqueous/gas equilibrium of aminium salts toward the gas phase. In comparison with the chlorides and nitrates of MA, EA, DMA, and PA, TMDEA sulfates are more stable under high T conditions. Additionally, high wind speeds enhance the emission of primary marine aerosols from sea spray and bubble bursting, further contributing amines in TSP, as amines are present in both seawater and primary marine aerosols. The source contributions and major secondary formation pathways of amines were summarized in Figure 5.

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4 Conclusions

This study presented a systematic analysis of the spatial variations, potential sources, and secondary formation mechanisms of low molecular weight amines in aerosols over the marginal seas of China. A consistent spatial pattern was observed for concentrations of total amines, water soluble inorganic ions, carbonaceous components, and > 100 organic compositions, exhibiting a north-to-south decreasing trend from the BS to the NYS and SYS. This pattern was attributed to the weakening influence of atmospheric pollutants transported from mainland East Asia, with the increasing contribution from the marine atmosphere.

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. MA, EA, and DMA were mainly derived from terrestrial biogenic and non-combustion anthropogenic sources, followed by fossil fuel combustion, with > 50% formed via nitrate-associated secondary formation pathways that interacted with BSOA formation. In comparison, PA mainly originated from combustion-related sources, along with terrestrial and marine biogenic sources, with a smaller fraction (35%) contributed by nitrate-associated secondary formation. Different from other amines, TMDEA was mostly (> 60%) generated through sulfate-associated secondary formation pathways, and was also contributed from primary marine aerosols, such as sea spray and bubble bursting.

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Terrestrial sources not only release gaseous amines but also provide atmospheric acidic species (e.g., SO_2 and NO_x), which participate in and accelerate new particle formation. Moreover, acidic aerosols transported from the mainland can further uptake gaseous amines from marine sources. These processes influence the physicochemical properties and climate effects of marine aerosols, as well as the carbon and nitrogen cycles. In addition to precursor abundance, ambient conditions, including T, RH, and atmospheric (photo-)oxidation capacity (e.g., OH, O_3 and NO_x concentrations), influence the secondary formation of amines, leading to temporal and spatial variations in the concentrations and compositions of amine in aerosols. Overall, these findings improve the understanding of amines in marine aerosols, highlight the significant impact of terrestrial emissions on offshore aerosol chemistry, and underscore the importance of multiphase chemical processes of amines under diverse ambient conditions.

Data availability. Data are available from the corresponding author on request (dryanlinzhang@outlook.com).

Supplement. The supplement related to this article is available online.

Author contributions. Xiao-Ying Yang wrote the draft and produced all the figures and tables. Fang Cao, Yu-Chi Lin, and Yan-Lin Zhang provided useful comments and revised the paper. Chang-Liu Wu, Yu-Xian Zhang, and Wen-Huai Song provided the measurement data.

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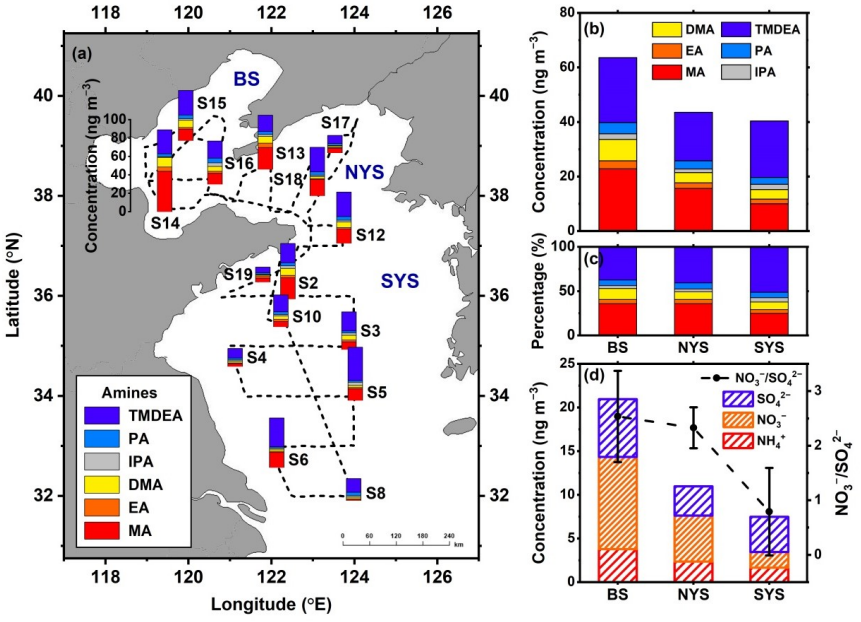

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941 **Figure 1.** Concentrations of amines in 15 TSP samples (a) collected along the cruise
942 track (black dotted line); average concentrations (b) and relative contributions (c) of
943 amines; and concentrations of NH_4^+, NO_3^-, and SO_4^{2-},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

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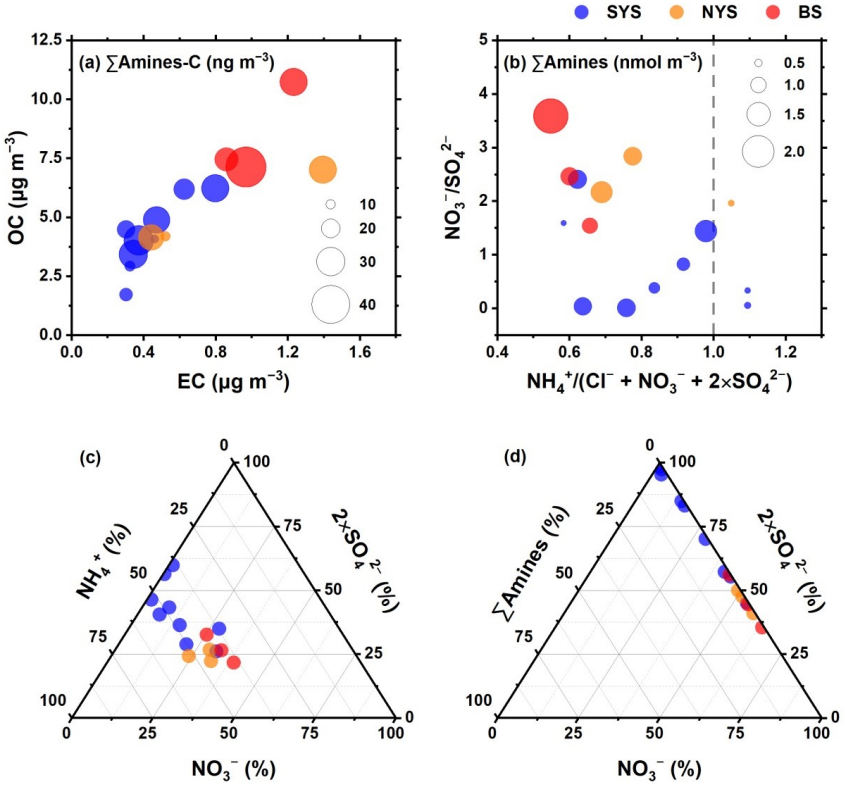

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947 **Figure 2.** Variations of ∑amines-C with OC and EC concentrations (a); variations of
948 ∑amines molar concentrations with the NO_3^-/SO_4^{2-} and $NH_4^+/(Cl^- + NO_3^- + 2\times SO_4^{2-})$
949 molar ratios (b); ternary diagram of the molar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

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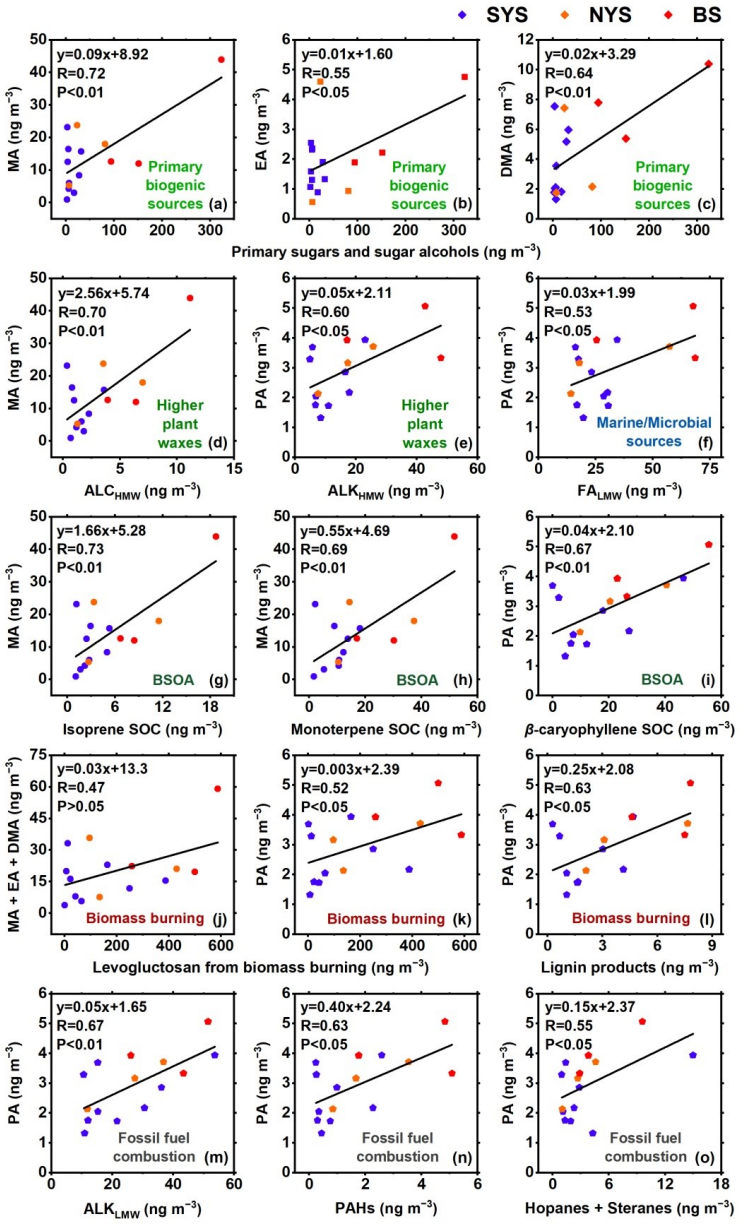

Figure 3. Linear regressions between amines and biomarkers (a–i), biomass burning tracers (j–l), and fossil fuel combustion tracers (m–o) in TSP over the SYS, NYS, and BS.

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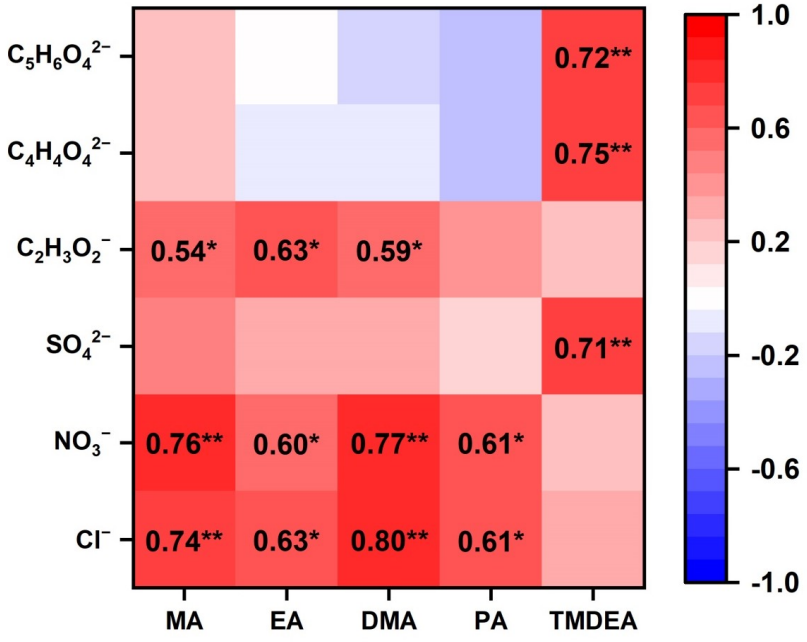

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959 **Figure 4.** Correlation coefficient matrix between amines and acidic species in TSP
960 over the YS−BS. Numbers indicate correlation coefficients that passed the
961 significance test; ** denotes $P < 0.01$, and * denotes $P < 0.05$.

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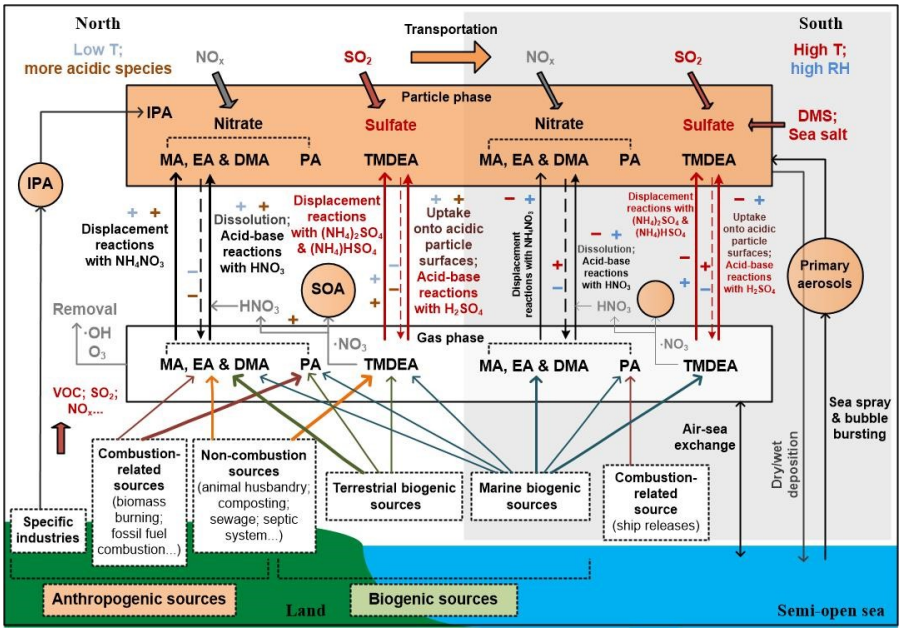

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964 **Figure 5.** Schematic diagram illustrating the source contributions and major
965 secondary formation mechanisms of amines, along with the influences of
966 environmental conditions over the YS−BS.

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